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TECHNICAL REPORT
76-64-FEL

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RESULT OF THE ASSAY OF RADIATION STERILIZED BEEF FOR INDUCED RADIOACTIVITY

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September 1976

UNITED STATES ARMY
NATICK RESEARCH and DEVELOPMENT COMMAND
NATICK, MASSACHUSETTS 01760

Food Engineering Laboratory

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- Radioassay

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Samples of beef sterilized by ionizing radiation were assayed using a NaI(Tl) detector and a multichannel analyzer to determine if measurable quantities of radioactivity had been induced during the irradiation process. The spectra obtained from each of two hundred sixty-one irradiated samples were compared with those obtained from 25 unirradiated controls, and no evidence of induced radioactivity was noted. Averaged spectra from irradiated samples, controls and the differences are presented in an appendix. Computer programs used in the data reduction are described briefly and the mechanisms of activation, if any, are reviewed.

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PREFACE

This report presents data in support of the U. S. Army's Radiation Preservation of Food Program. Calculations based on fundamental physical principles have shown that, where energy considerations permit, amounts of radioactivity induced in beef during radiation sterilization would be very small. Results presented in this report indicate experimental agreement with these theoretical calculations. The authors would like to express their appreciation to Mr. John Sieckarski for his help in the counting of these samples.

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RESULT OF THE ASSAY OF
RADIATION STERILIZED BEEF
FOR INDUCED RADIOACTIVITY

by

Thomas G. Martin, III
and
Robert L. Becker

Introduction

In order to establish the wholesomeness of beef sterilized by ionizing radiation, an animal feeding study of long duration was undertaken. In support of this study 96,000 kg of enzyme-inactivated (EI) beef were procured in seven batches over a period of approximately three years. One half was irradiated with gamma rays from a megacurie ^{60}Co source and the other half was irradiated with 10 MeV electrons from a linear accelerator (linac). Beef samples received doses ranging from 4.6 to 7.1 Mrad and were maintained in a frozen condition during the irradiation.

The study reported here, undertaken concurrently with the animal feeding study, was designed and conducted to determine whether radioactivity is induced in meat samples at a measurable level during the irradiations, and to demonstrate a procedure that could, if necessary, be adopted for routine monitoring in production-type facilities. Gamma ray spectra from irradiation-sterilized beef were obtained during counts of long duration (6×10^4s) with a detector system of high efficiency. The results indicate that no measurable radioactivity is induced either by ^{60}Co gamma radiation or by 10 MeV electrons. In addition, a preliminary theoretical analysis of the mechanisms of radioactivity production is presented.

Experimental Procedure

Upon receipt of each batch of EI beef, samples of meat packaged for either ^{60}Co gamma or linac electron irradiation (Fig. 1) were selected at random to serve as unirradiated controls of the batch that they represented. These samples were kept frozen until just prior to preparation for counting.

Approximately 350 kg of meat were assayed, comprising 261 irradiated samples and 25 controls. Immediately prior to counting, each sample was ground, transferred to a Marinelli beaker, weighed, and placed on the 7.62 cm x 7.62 cm NaI(Tl) crystal of a scintillation detector for counting. The detector was surrounded by a 10-cm Pb shield.

Figure 2 is a block diagram of the instrumentation used in the radioassay. The primary components are the NaI(Tl) scintillation counter, a Nuclear-Chicago 1600-Channel pulse height analyzer, a Tally paper tape punch, a Monroe lister, and a Houston Omnigraph Plotter. The Marinelli beaker provides a means to surround the scintillation crystal with meat sample. No portion of the sample was more than 10 cm from the crystal.

Each sample was counted for 1000 minutes. Visual inspection of the CRT display was made before dsta were removed from the spectrometer to determine if any unusual peaks or significant increase in counts above thos obtained from control samples were obtained from any irrsdiated sample. All spectrs were recorded by the Tally punch, the Monroe lister, and a Houston Omnigraphic plotter, and then were transferred from paper tape to msgnetic tape to allow analysis on the U. S. Army Nstick Research and Development Command Univsc 1106 computer.

Efficiency of the counting system was determined by the counting of several standard solutions in the same configurstion as was employed for meat samples, with water substituted for ground beef. The radionuclides used included ^{57}Co, ^{203}Hg, ^{137}Cs, ^{22}Na, snd ^{40}K. Figure 3 is a plot of this calibration.

Analysis of Data

The most distinctive photopeaks observe in each spectrum are from the naturally-occuring gamma rays st 0.511 MeV (the positron annihilation energy) and 1.460 Mev (^{40}K gamma ray). These were used as reference peaks to correct the energy scale for drift of the spectrometer.

The computer was used to search through esch spectrum, analyzing small portions at a time, to locate individual peaks from rsdionuclides that might be present in the samples or which might contribute to the room background. Several such peaks were found, snd all were identified as naturally-occuring radiations, with the exception of ^{137}Cs which results from fallout from testing of nuclear weapons.

As a further check on the possibility that induced radioactivity might be present in the samples, an averaged gsmma spectrum was obtained from each batch of linac electron irradiated meat as well as from each batch of ^{60}CO gamma ray irradisted meat. Then the gsmma spectrum for the unirradiated control sample of each group of samples wss subtracted from the corresponding averaged spectrum. In Appendix A the aversged spectrum, control spectrum, and difference spectrum are presented for esch group of samples.

The difference spectrum in each csse is observed to be very nearly zero. Small deviations from zero are attributable to statistical variations because of the random arrivsl of gamma rays st the detector, or occasionally because a photopeak in the averaged spectrum may be brosder than the corresponding peak in the control. There is no evidence of a residusl effect that could be interpreted as either a photopeak from s gamms emitter or a bremsstrahlung spectrum from beta-activity in the mest sample.

A brief discussion of esch computer progrsm employed in this data analysis is given in Appendix B.

Discussion

The processes by which radionuclides can be induced in food samples during sterilization by ^{60}Co gamma rays or 10 MeV electrons are well known. They include isomeric excitation, which would be possible for either source, and electronuclear reactions and neutron activation, which are impossible to produce by ^{60}Co gamma rays and are very unlikely processes with 10 MeV electrons.

It is the nature of isomeric excitation that those states with longer half-lives have the smaller probability for excitation. Furthermore, those states for which the lifetime exceeds a few seconds are in elements (e.g., In, Sn, and Cd) which are present in meat in only trace amounts, if at all.

On the basis of measurements made at this laboratory, as well as other data, we expect the greatest isomeric excitation in meat to occur in the 14-day state of ^{117}Sn. Our yield factor for the production of ^{117m}Sn is measured to be 1.5 μCi/kg$\cdot$ Mrad (5.5×10^4 Bq/kJ) which agrees well with the data of Glass and Smith.[1] Taking into account the 7.6% isotopic abundance of ^{117}Sn and the elemental abundance of Sn in meat (approximately 1.0 ppm), we calculate an isomeric yield at 6 Mrad of 0.66 pCi/kg (0.024 Bq/kq). This is about one ten-thousandth the average measured activity of ^{40}K occurring naturally in meat and is far below our ability to measure.

The thresholds for most photonuclear or electronuclear reactions are above the electron nergies which are employed in the irradiation of meats. Two exdeptions would be $^{2}H(\gamma,n)\,^{1}H$ and $^{13}C(\gamma,n)\,^{12}C$, but these reactions produce stable nuclides. Generally, the cross sections for those reactions which are energetically possible are so low over the energy range between threshold and 10 MeV that no significant activation results. However, as discussed below, it is possible that neutrons from (γ,n) reactions, particularly from ^{2}H or ^{13}C, might themselves produce subsequent activations.

For neutron activation to occur, the neutrons produced by (γ,n) or (e,n) reactions in ^{2}H or ^{13}C will be slowed down in the meat sample, then be captured by an element in the meat, for example Na, Cl, P, or Mn.

Isotopic ahundances of ^{2}H and ^{13}C are very low (0.01 and 1.1 percent, respectively), end the shape of the meat boxes used in linac irradiation does not provide efficient thermalization or slowing down of the neutrons. Neither Mn or Cl activation in the meat samples was observed. It is not clear whether Na activation occurred, since the ^{24}Na gamma ray photopeak which would result would be obscured by that of ^{40}K which has nearly the same

[1]Glass, R. A. and H. D. Smith, Radioactive Isomer Production in Foods by Gamma Rays and X-Rays. Stanford Research Institute Report to U. S. Army Quartermaster Research and Engineering Command, Report No. 3 (Final) October 3, 1960.

energy. However, attempts to observe Na activation by implanting 10 g samples of NaCl in meat, then removing them for counting, have been unsuccessful so far, although they are continuing.

Interpretation of the Gamma Spectra

The calibration data of Fig. 3 provide a means of determining the activity in the meat sample of a nuclide for which a photopeak is observed, as well as of determining an upper limit for the activity of a nuclide for which a photopeak is not seen.

The large concentration of potassium normally present in beef allows a check on the reliability of this calibration through the detection of the 1,460 MeV gamma ray from ^{40}K. Our calibration studies determined that the detection system has an efficiency of 2740 counts/kg (K) min for ^{40}K, while the measured spectra gave the order of 4.7 counts/kg (beef) min in the ^{40}K photopeak.

This corresponds to a ^{40}K activity in the meat samples of 3600 disintegration/kg min or 1600 pCi/kg (59.2 Bq/kg). Using the known isotopic abundance (118 ppm) and lifetime (1.27×10^9 yr), we determine an elemental abundance for potassium of 0.0020. Since this is in reasonable agreement with values typically found to be about 0.0033,[2] we conclude that our procedures are correct.

Cesium-137 was found in measurable levels only in the earlier batches. The samples counted in 1973 and 1974 were below 10 pCi/kg (0.37 Bq/kg) levels; however earlier samples were as high as 50 pCi/kg (1.85 Bq/kg). These data agree with the results published by Simpson et al.

The results of this study demonstrate that no photopeaks that may be attributed to induced radioactivity can be observed in gamma-ray spectra from radiation sterilized beef. Furthermore, no residual effect of irradiation can be seen by subtracting any control sample from the averaged spectra of irradiated samples of that batch. Thus the induced radioactivity if present was unmeasurable, and if present it yielded a gamma flux smaller than that of the background radiation from the environs, which was the limiting factor in our sensitivity. We conclude that any induced radioactivity must be smaller than 0.1 percent of the radioactivity that is normally present in beef samples.

[2] Lawrie, R. A., Meat Science, 2nd Edition, p. 349, Pergamon Press, (1974).

[3] Simpson, R. E., E. J. Baratta, and C. F. Jelinek, Radionuclide in Food: Monitoring Program, Radiation Data and Reports Vol. 15, No. 10, October 1974.

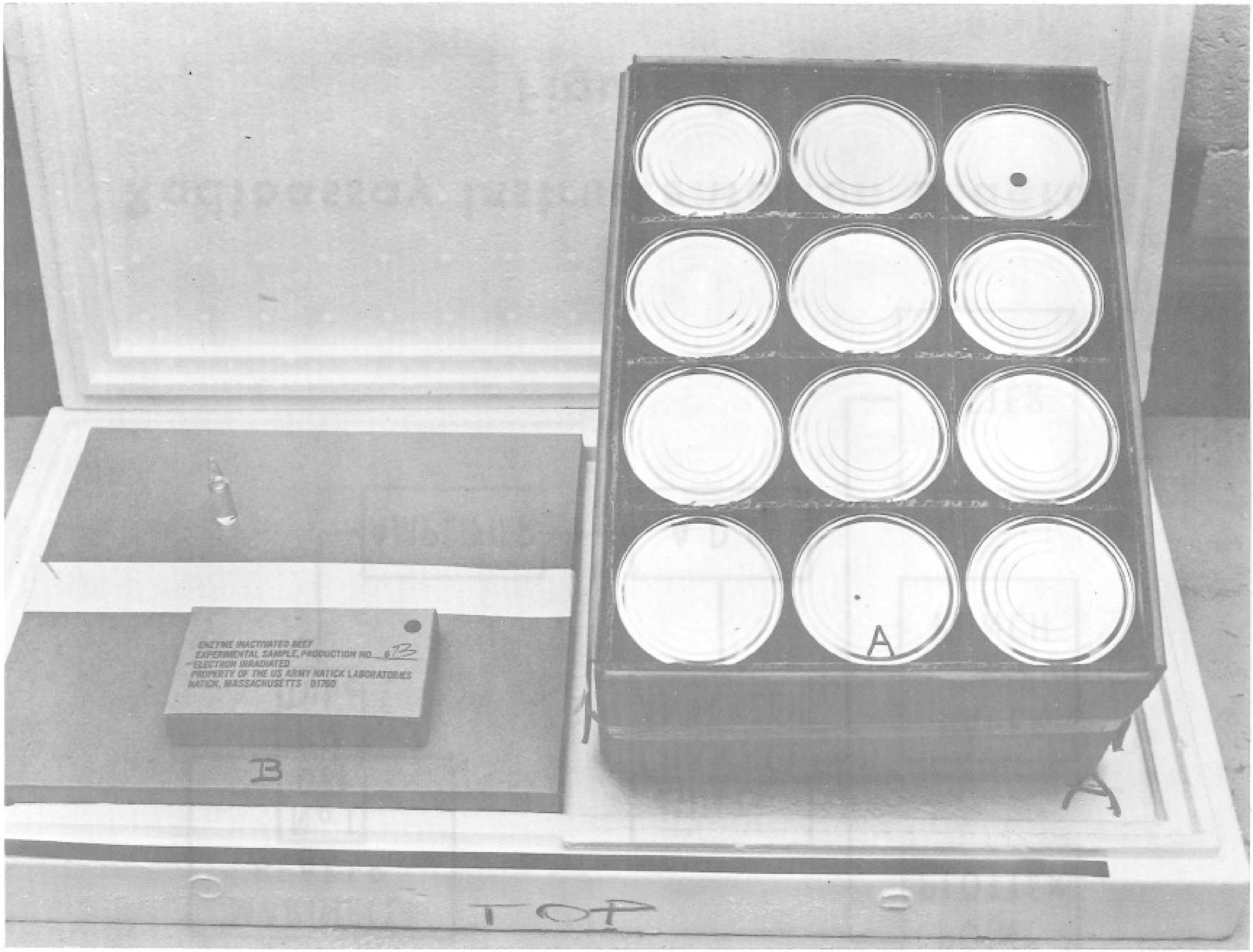
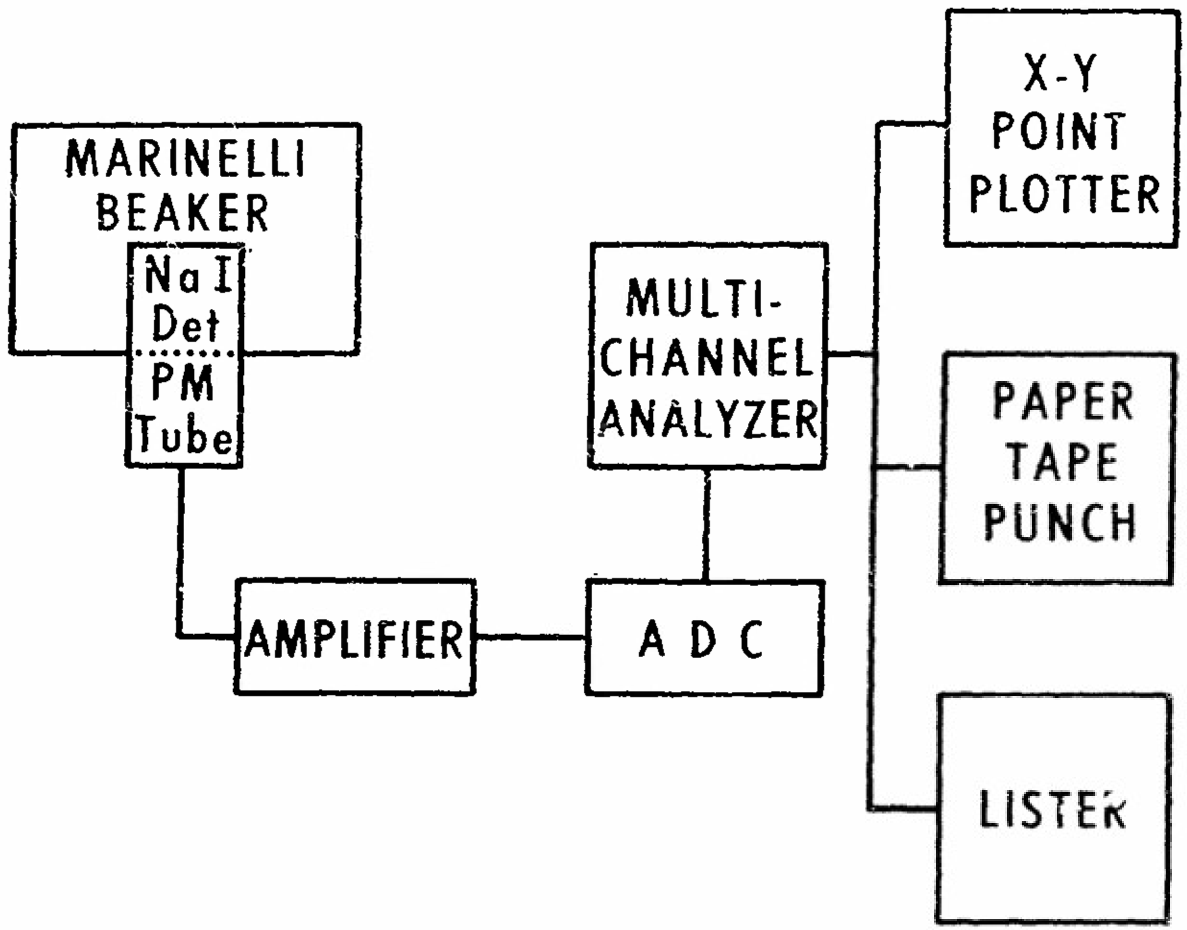


FIGURE 1

BEEF PACKAGED FOR IRRADIATION BY COBALT-60 GAMMA RAYS (A)
OR BY 10 MeV ELECRONS (B)



Radioassay Instrumentation Diagram

Figure 2

Photopeak Efficiency for 7.62x7.62cm NaI (Tl) crystal and a 3 liter Marinelli Beaker

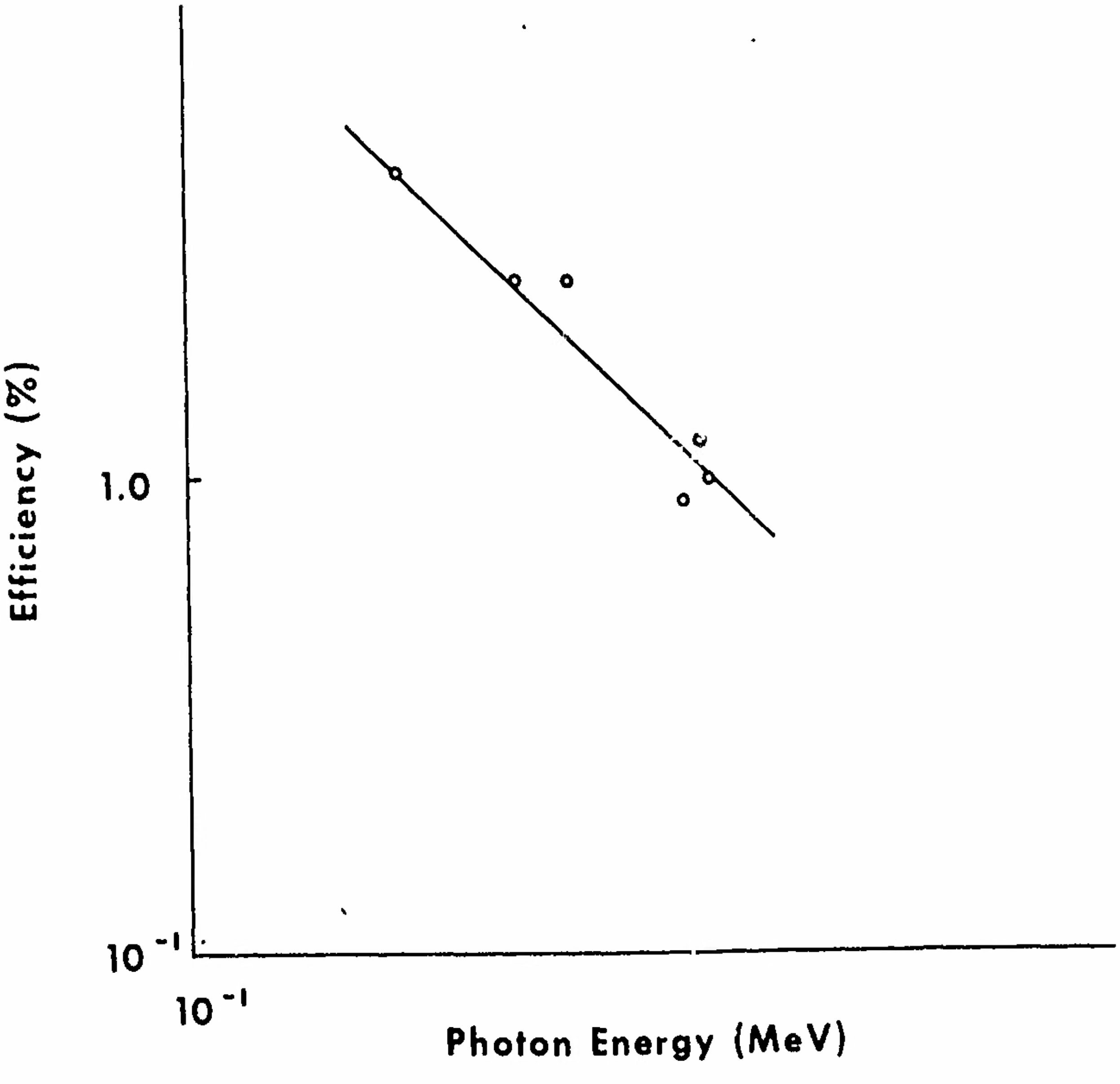
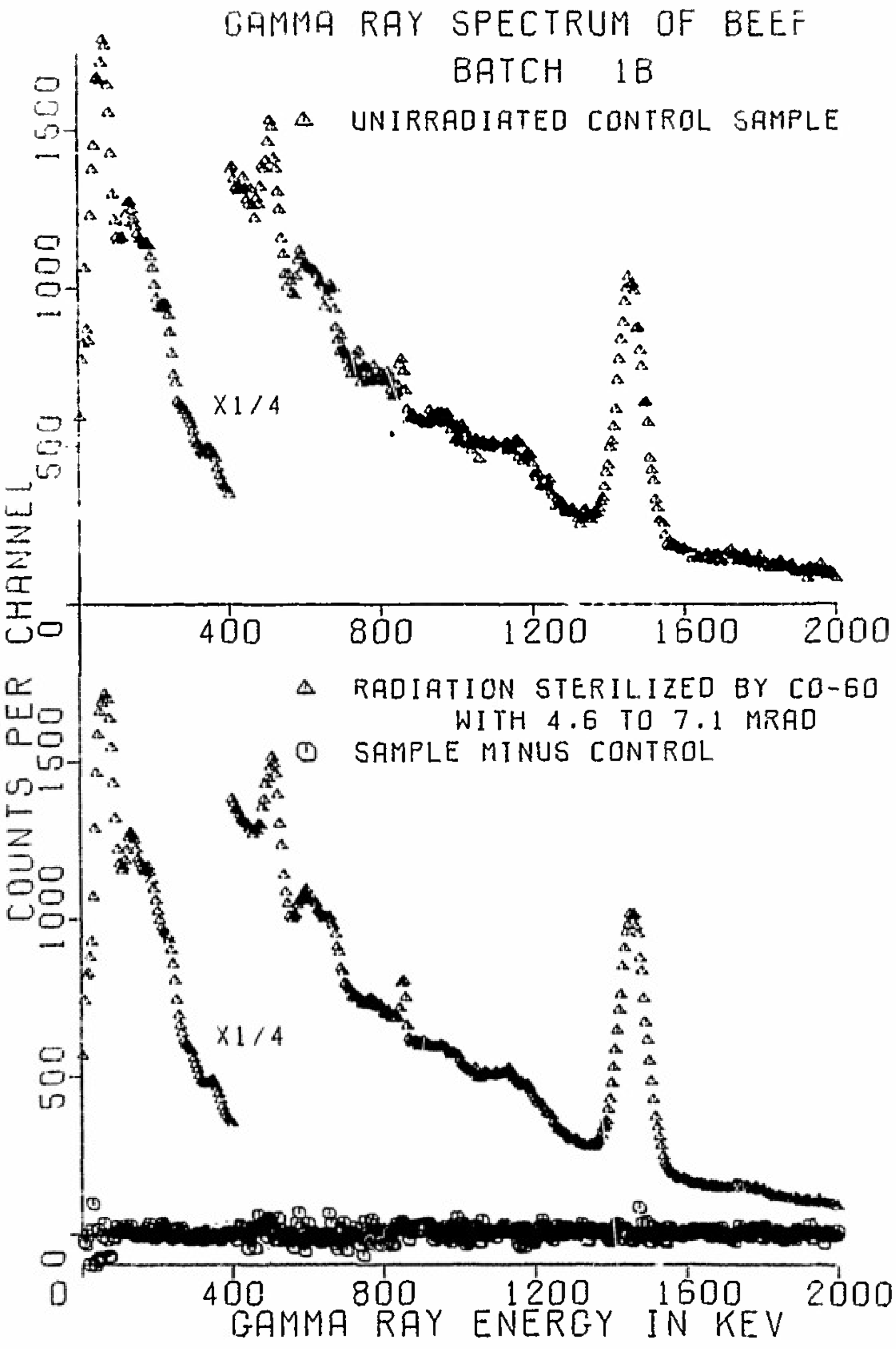


Figure 3

APPENDIX A

GAMMA RAY SPECTRA OF BEEF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BATCH 1B
UNIRRADIATED CONTROL SAMPLE
X1/4
RADIATION STERILIZED BY CO-60
WITH 4.6 TO 7.1 MRAD
SAMPLE MINUS CONTROL
X1/4
COUNTS PER CHANNEL
GAMMA RAY ENERGY IN KEV
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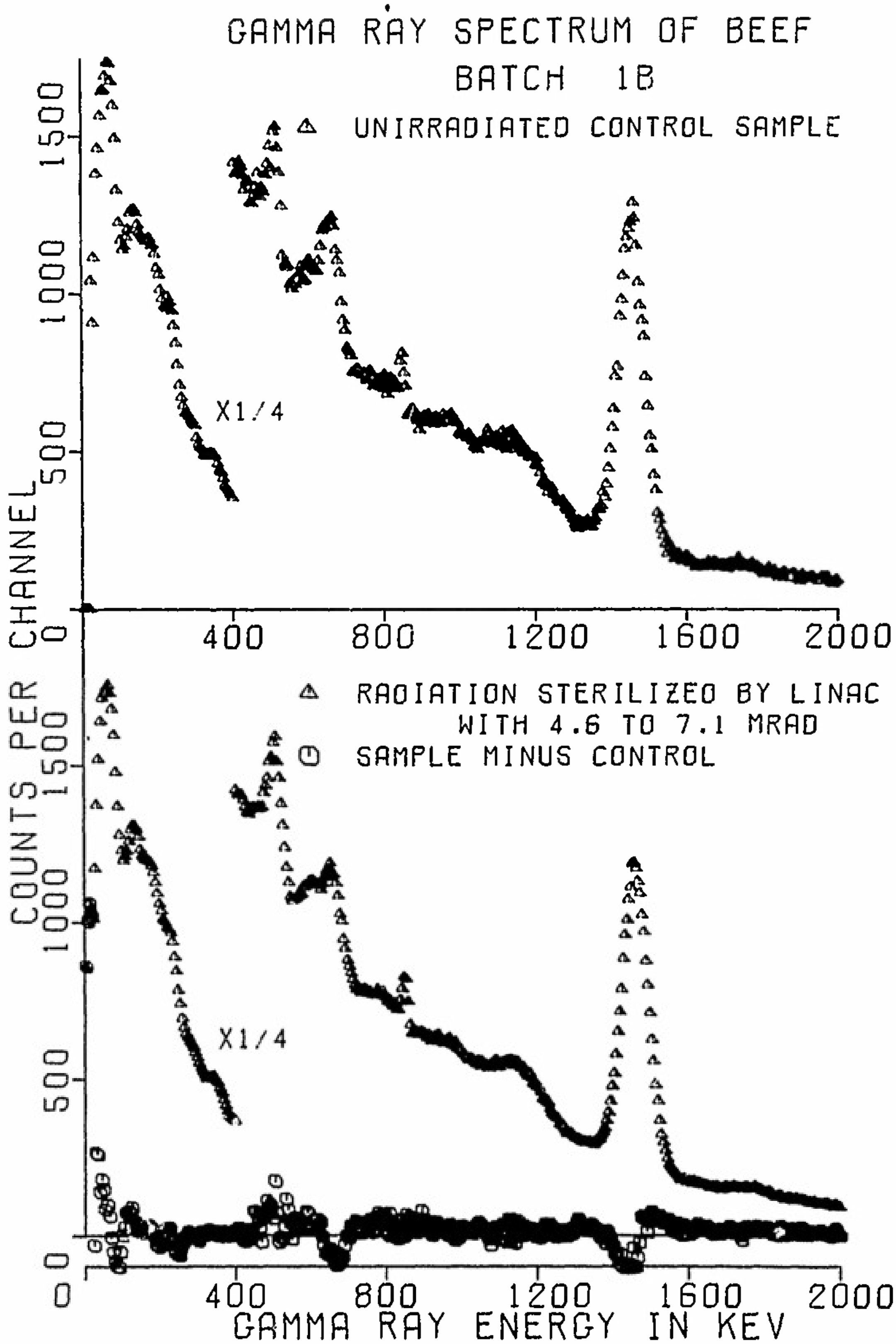
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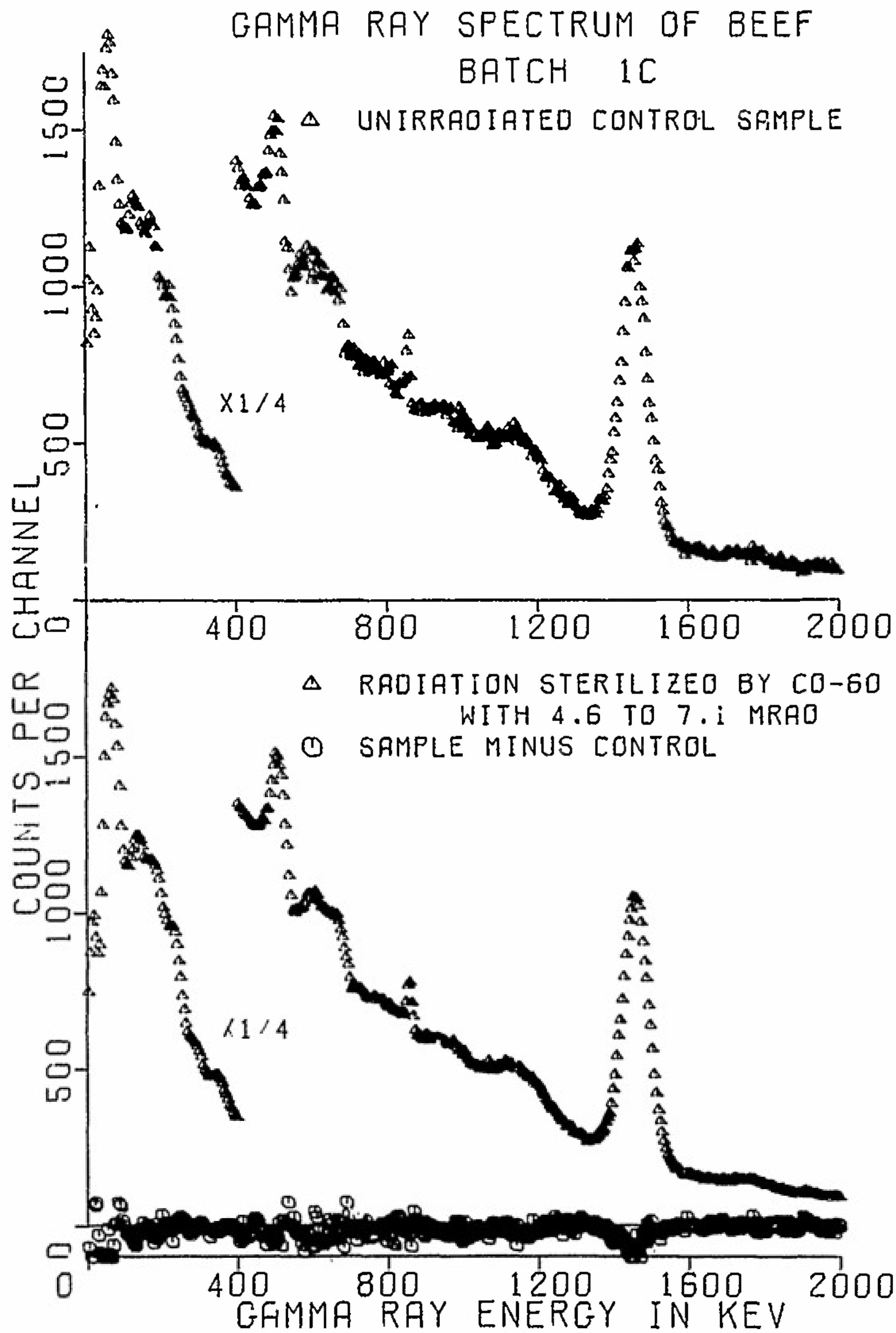
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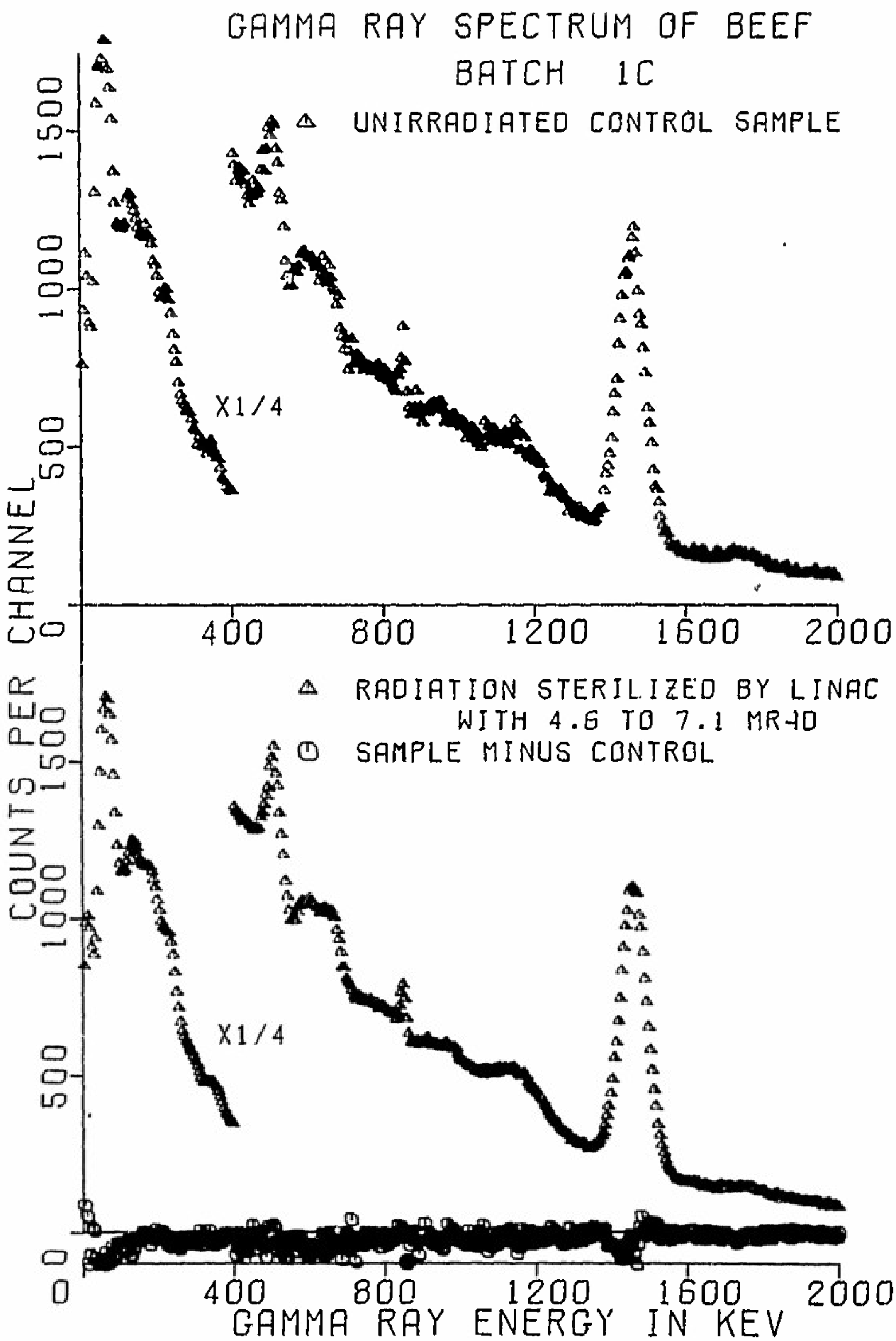
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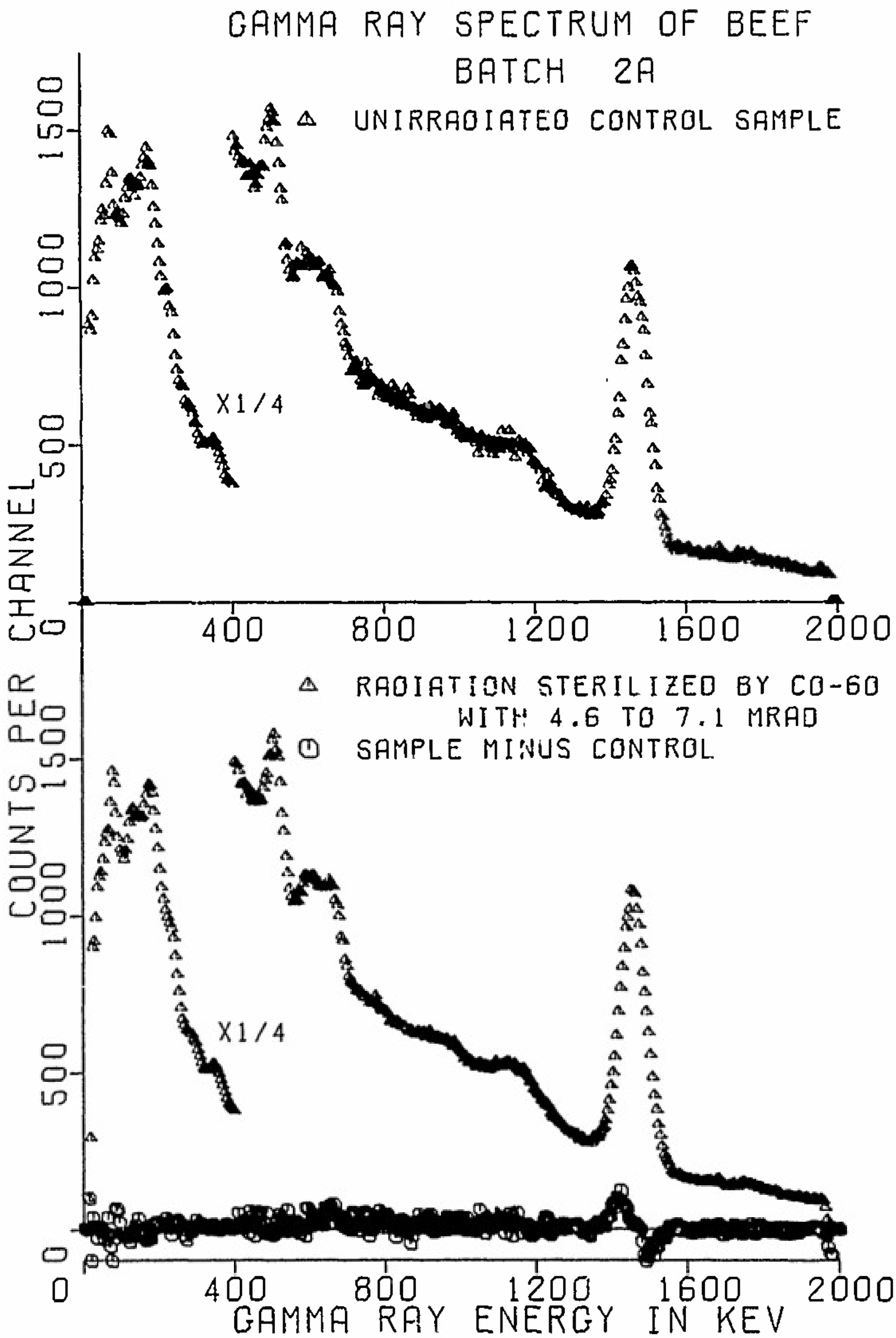

GAMMA RAY SPECTRUM OF BEEF
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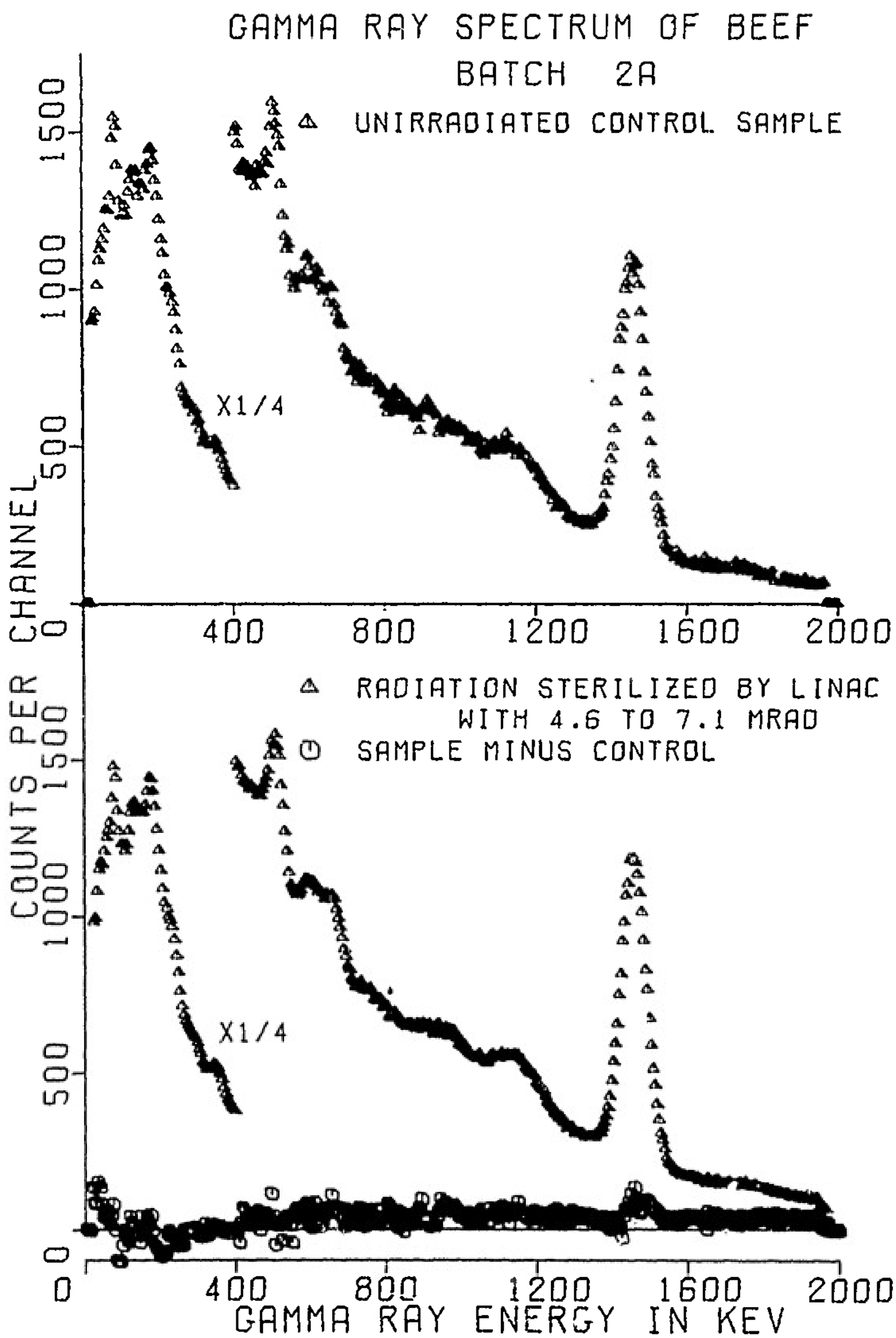
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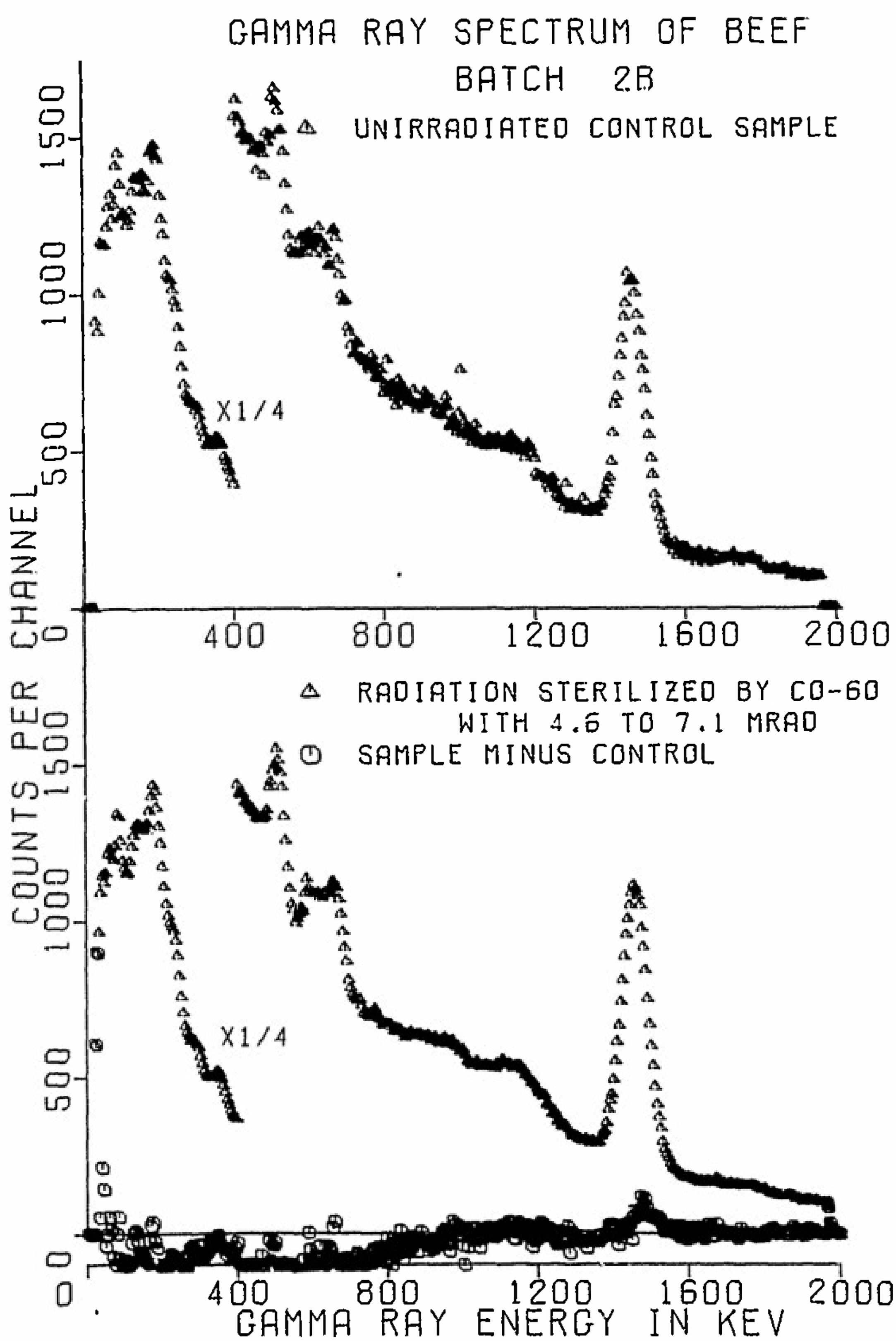
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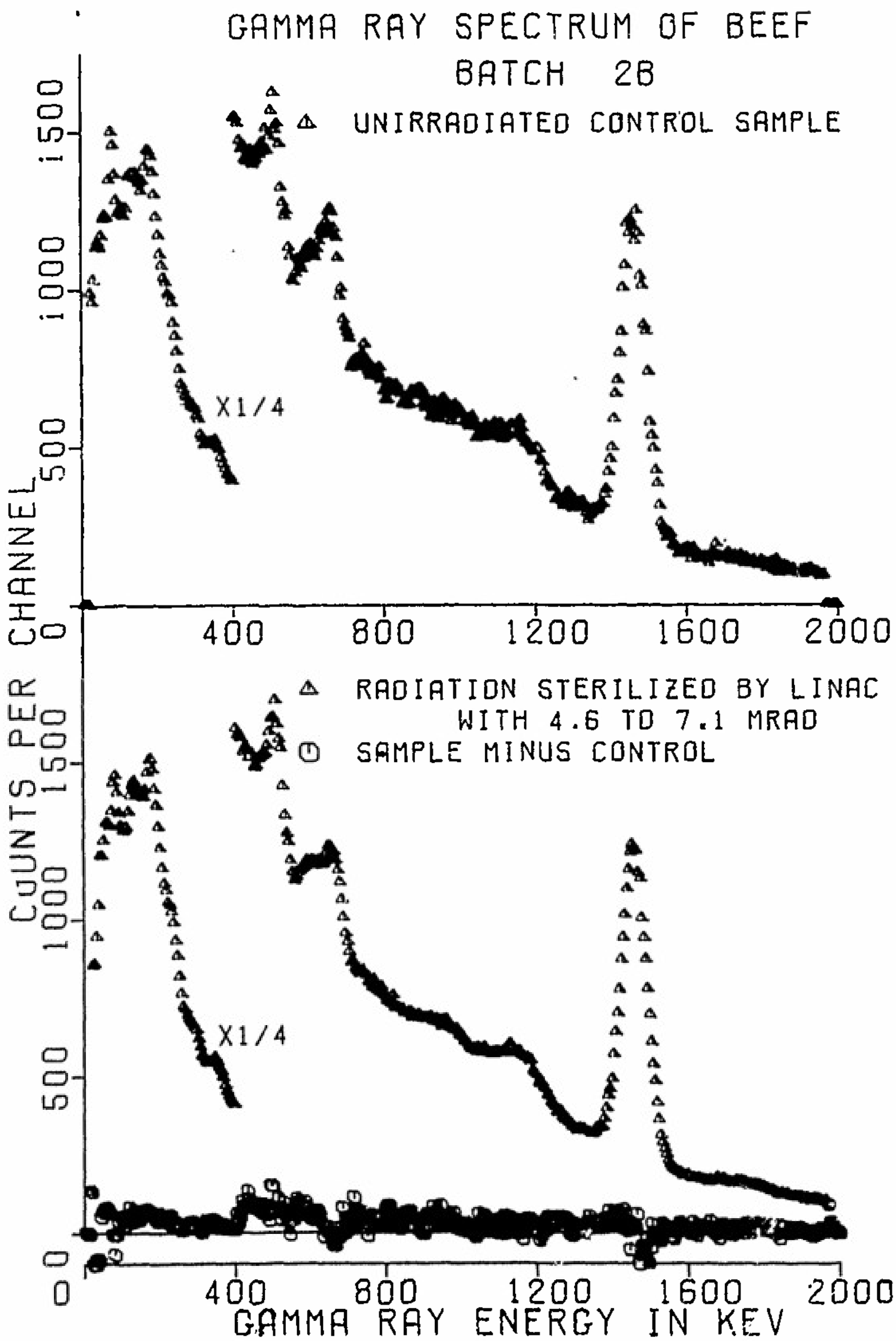
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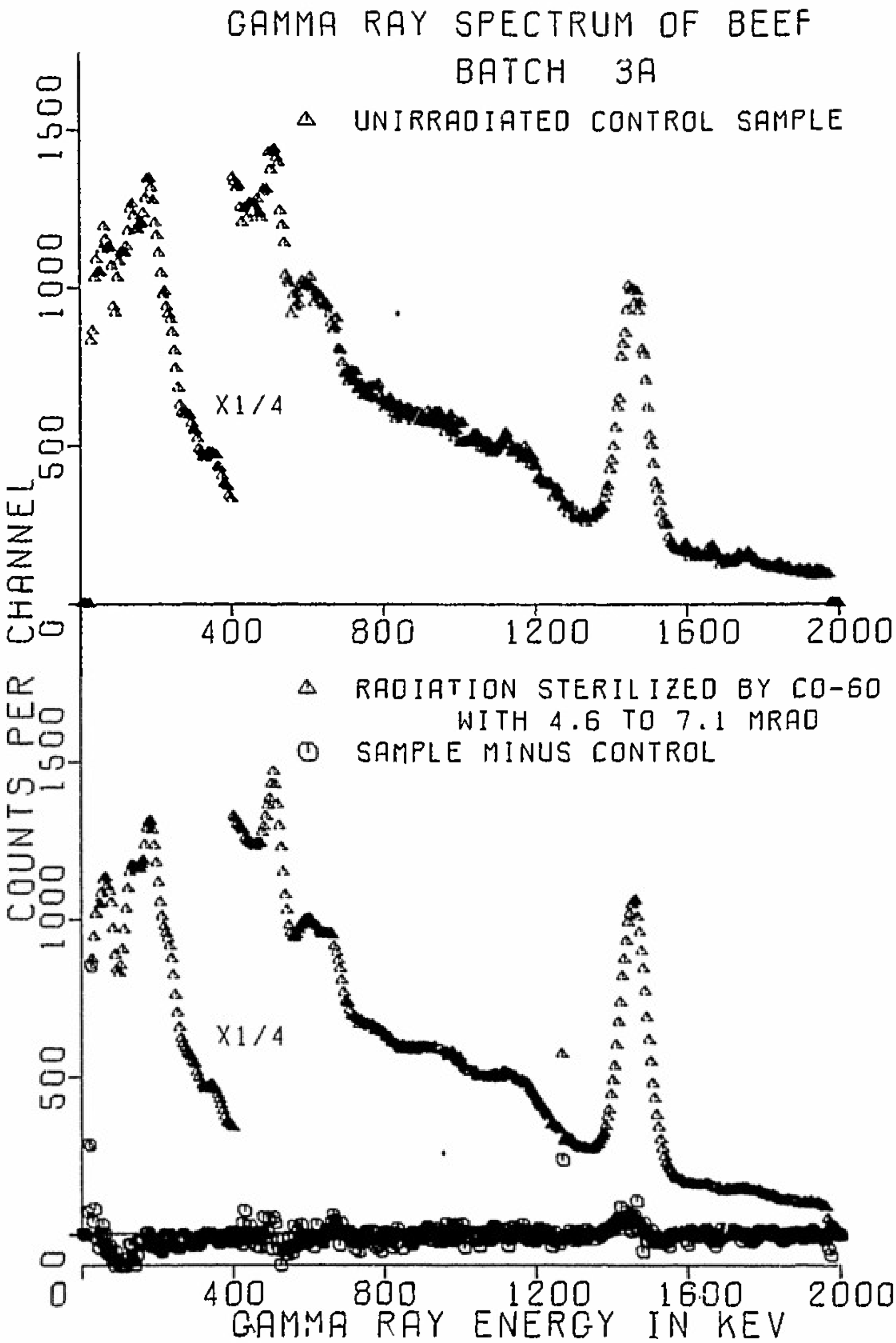
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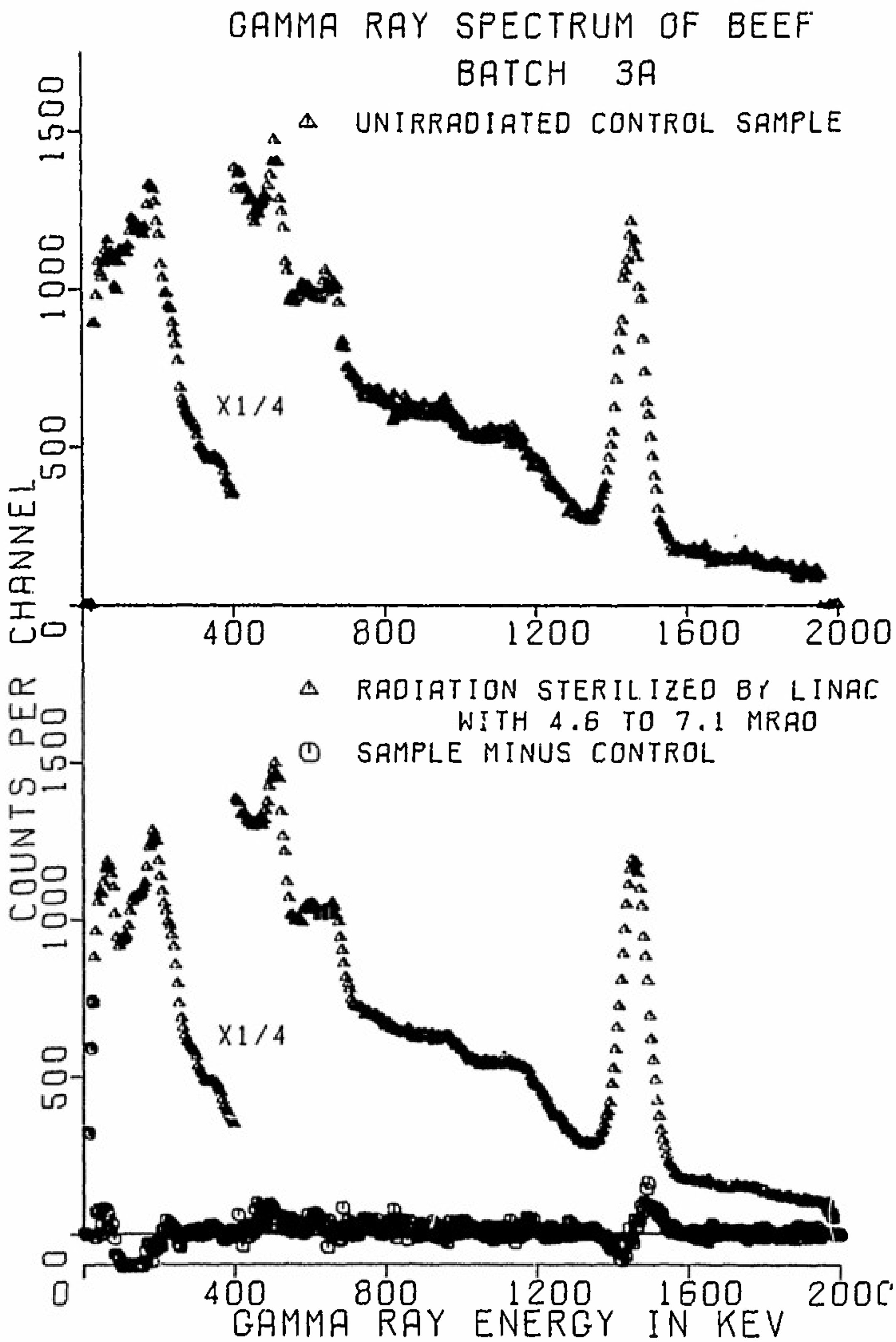
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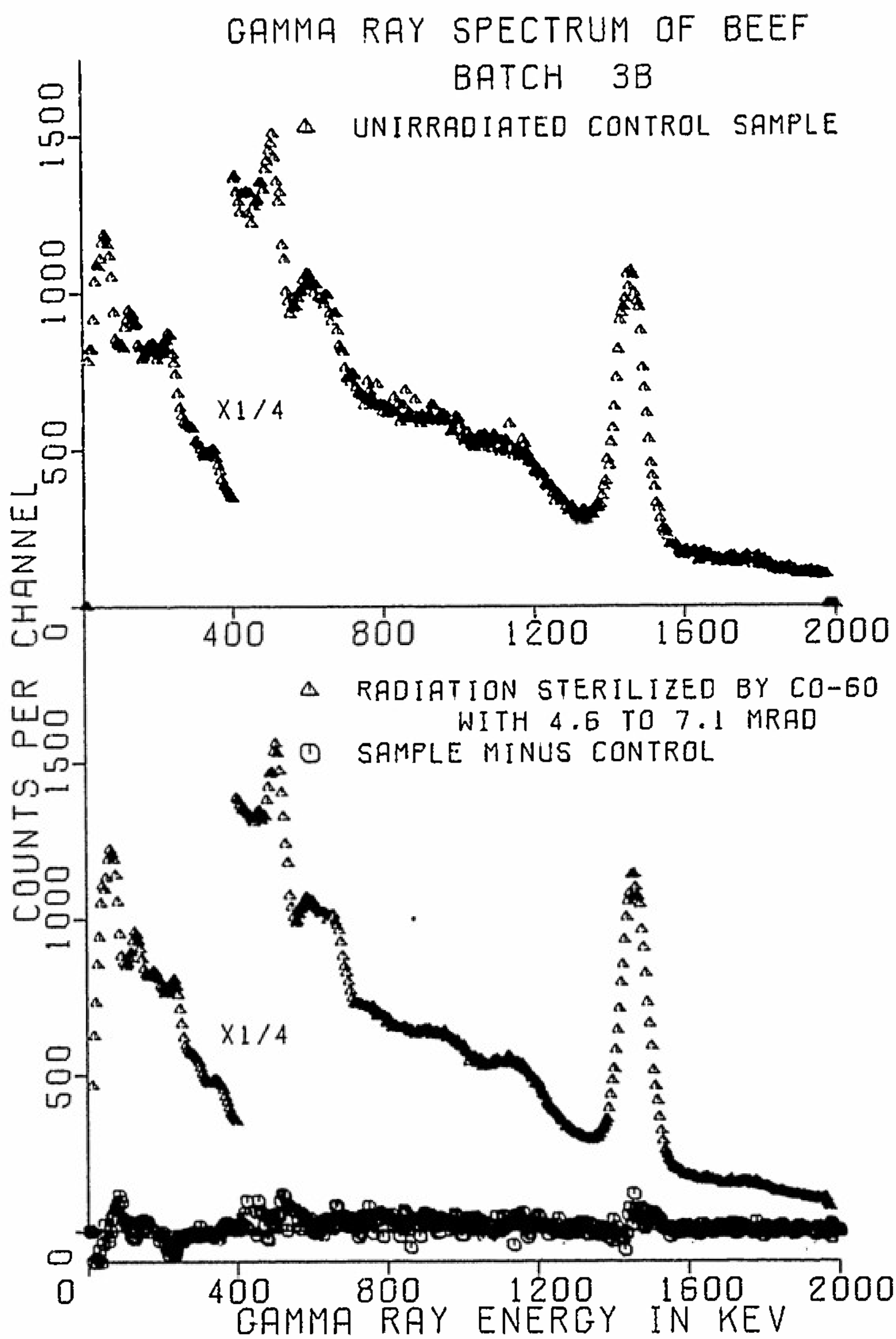
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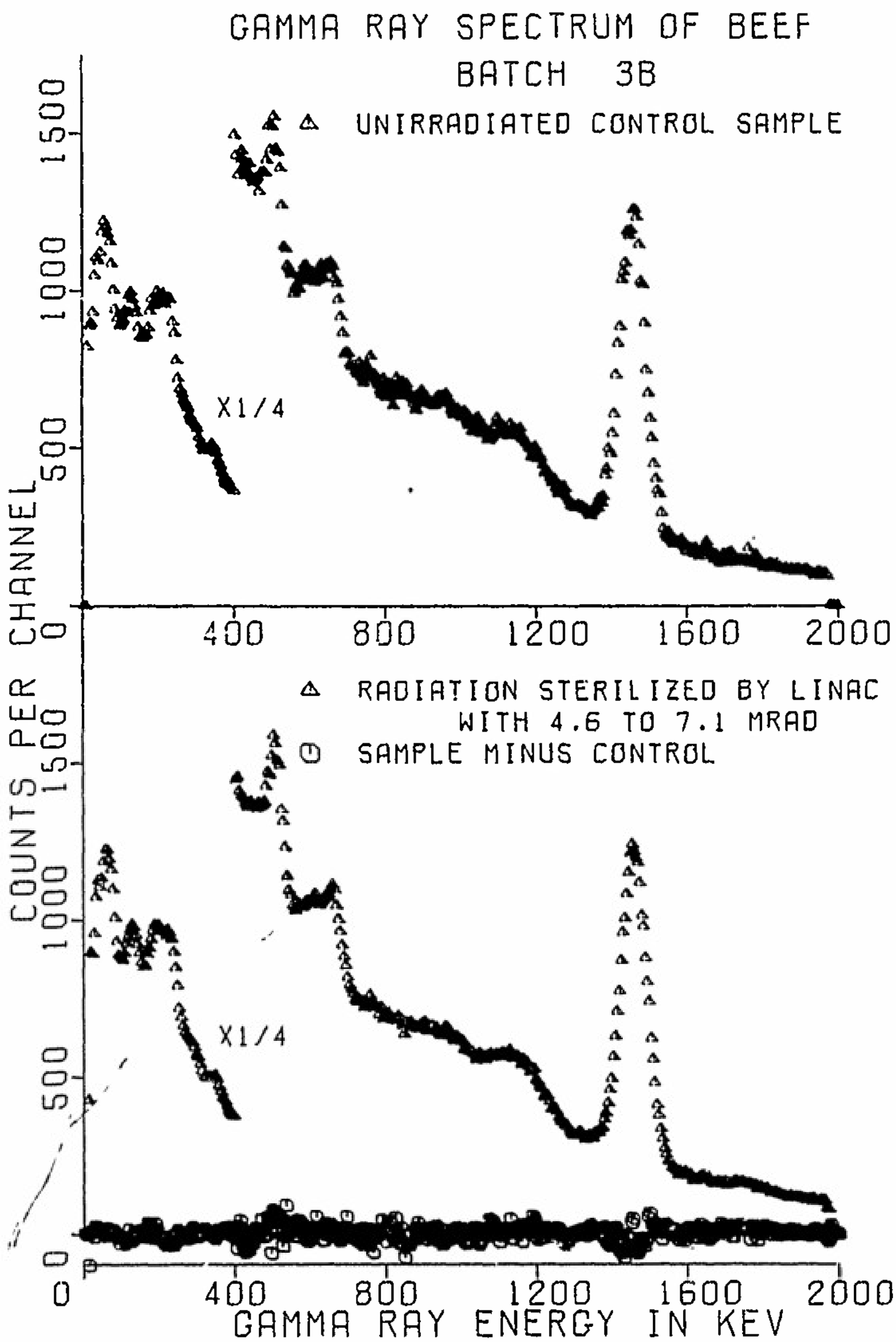
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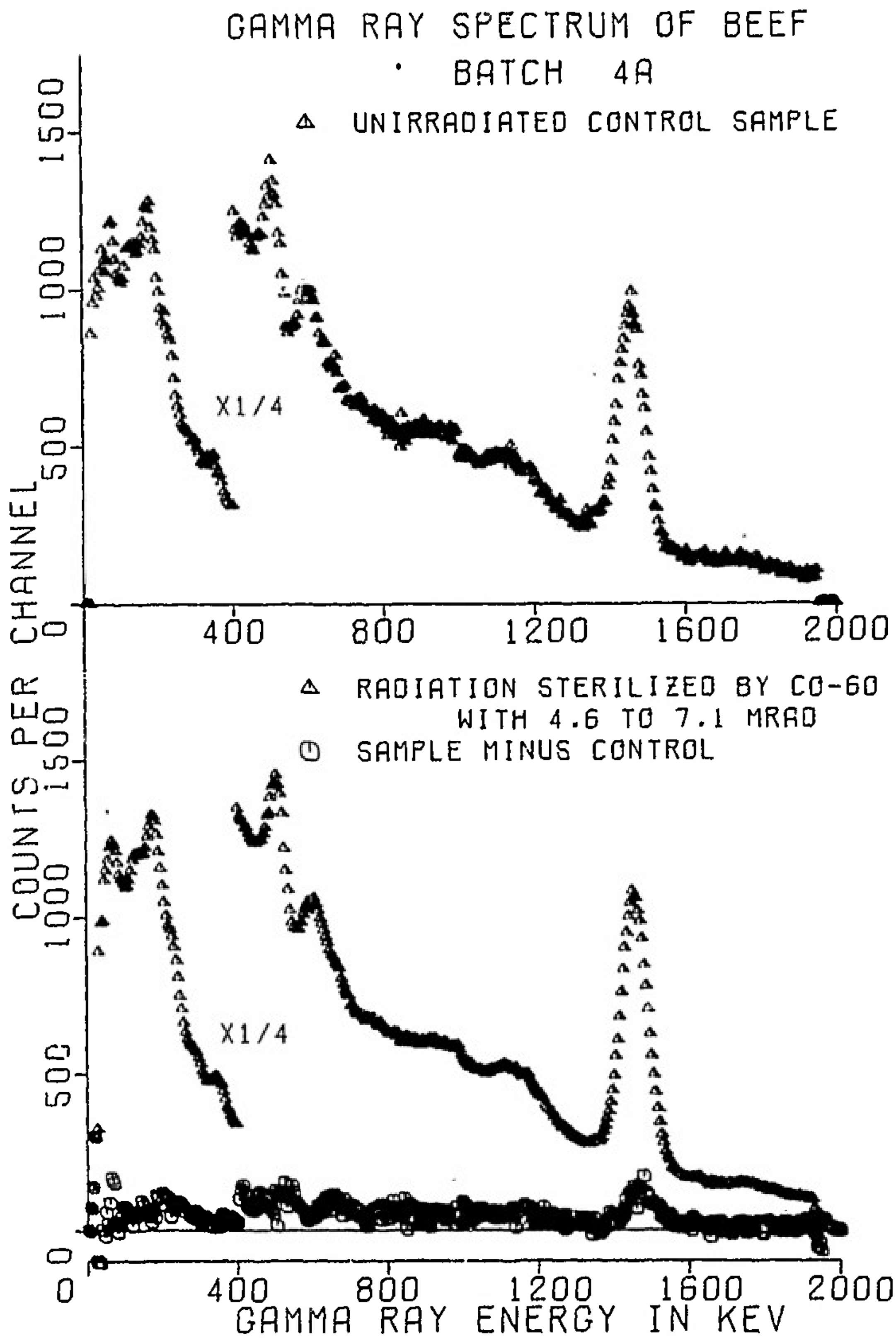

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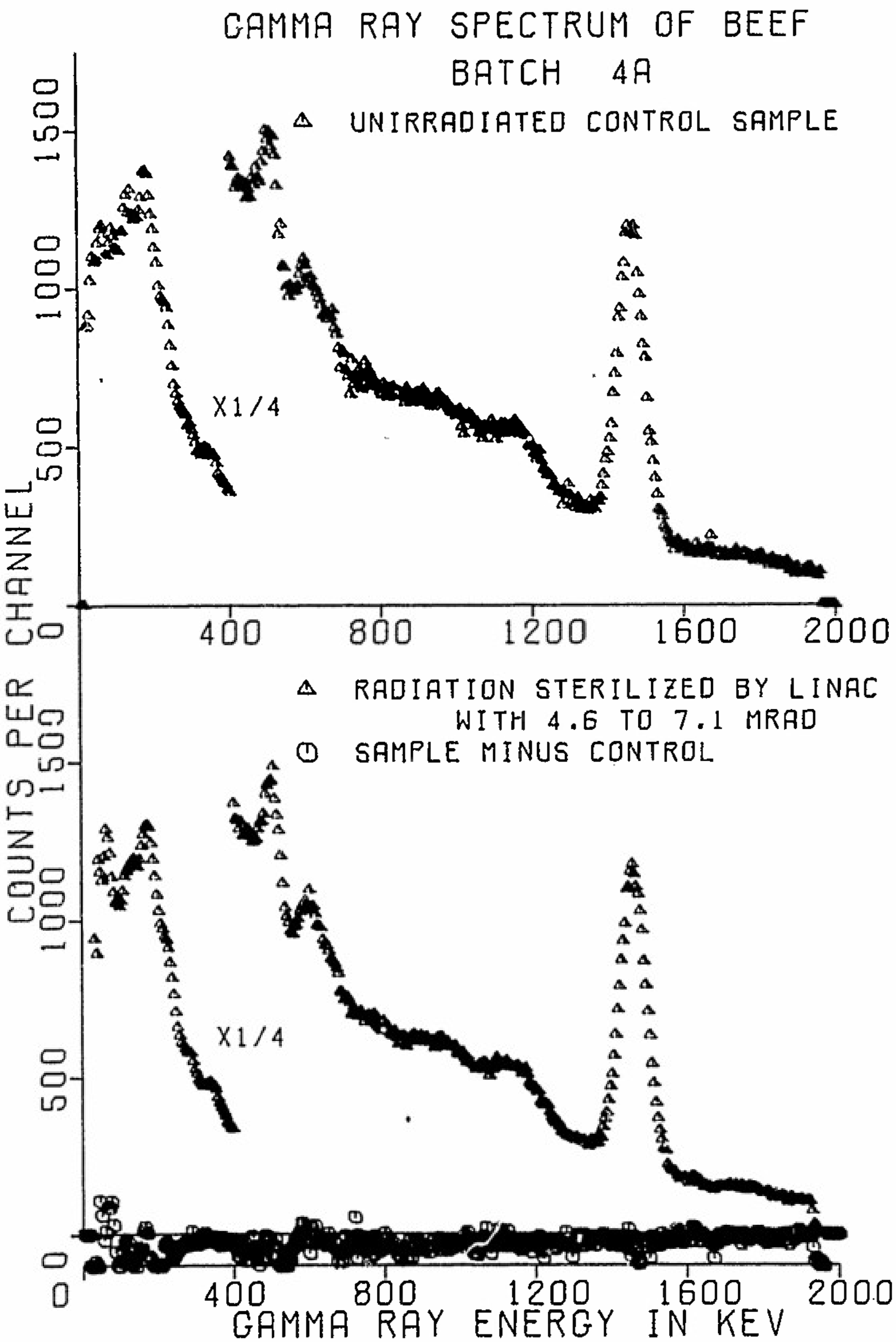
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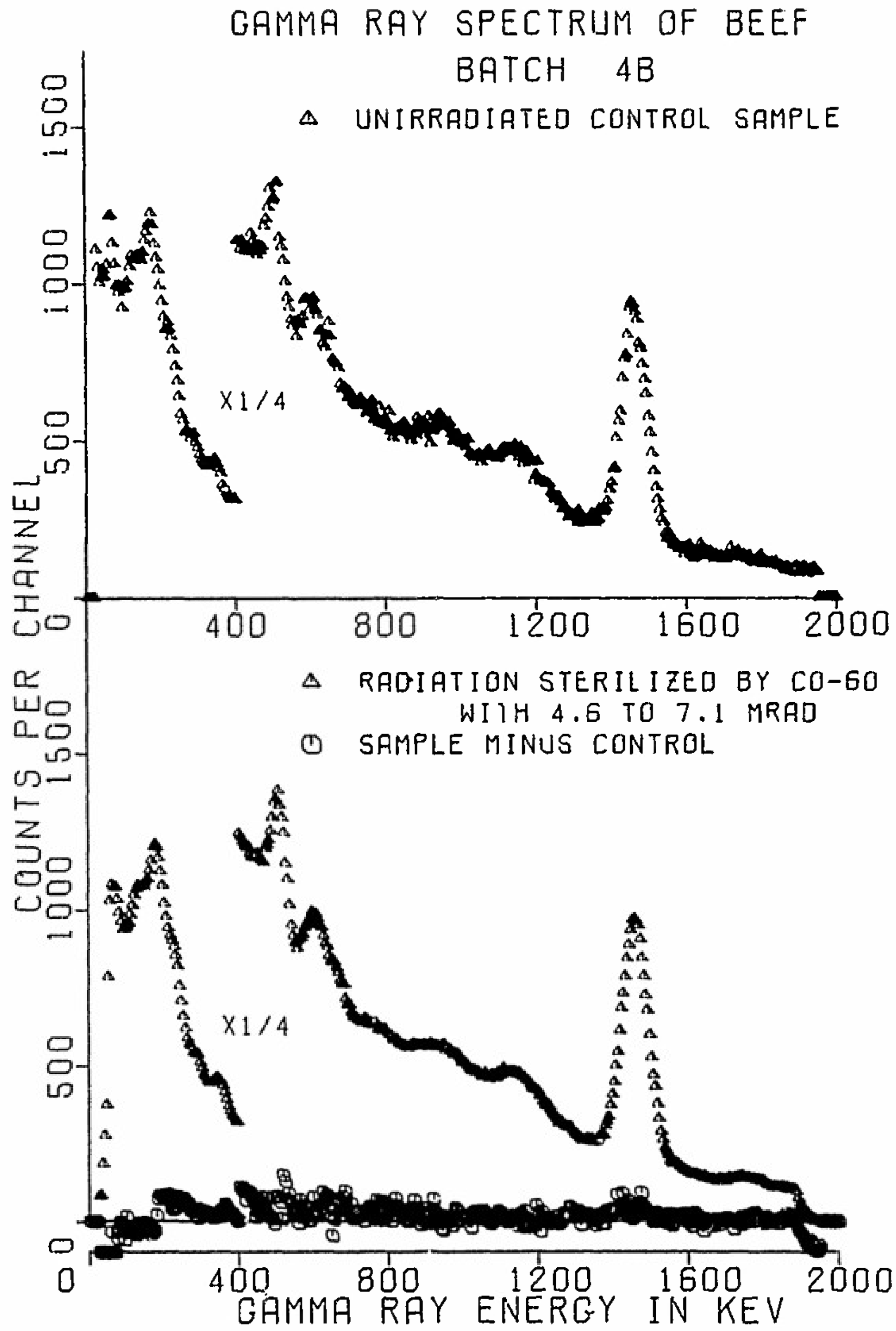
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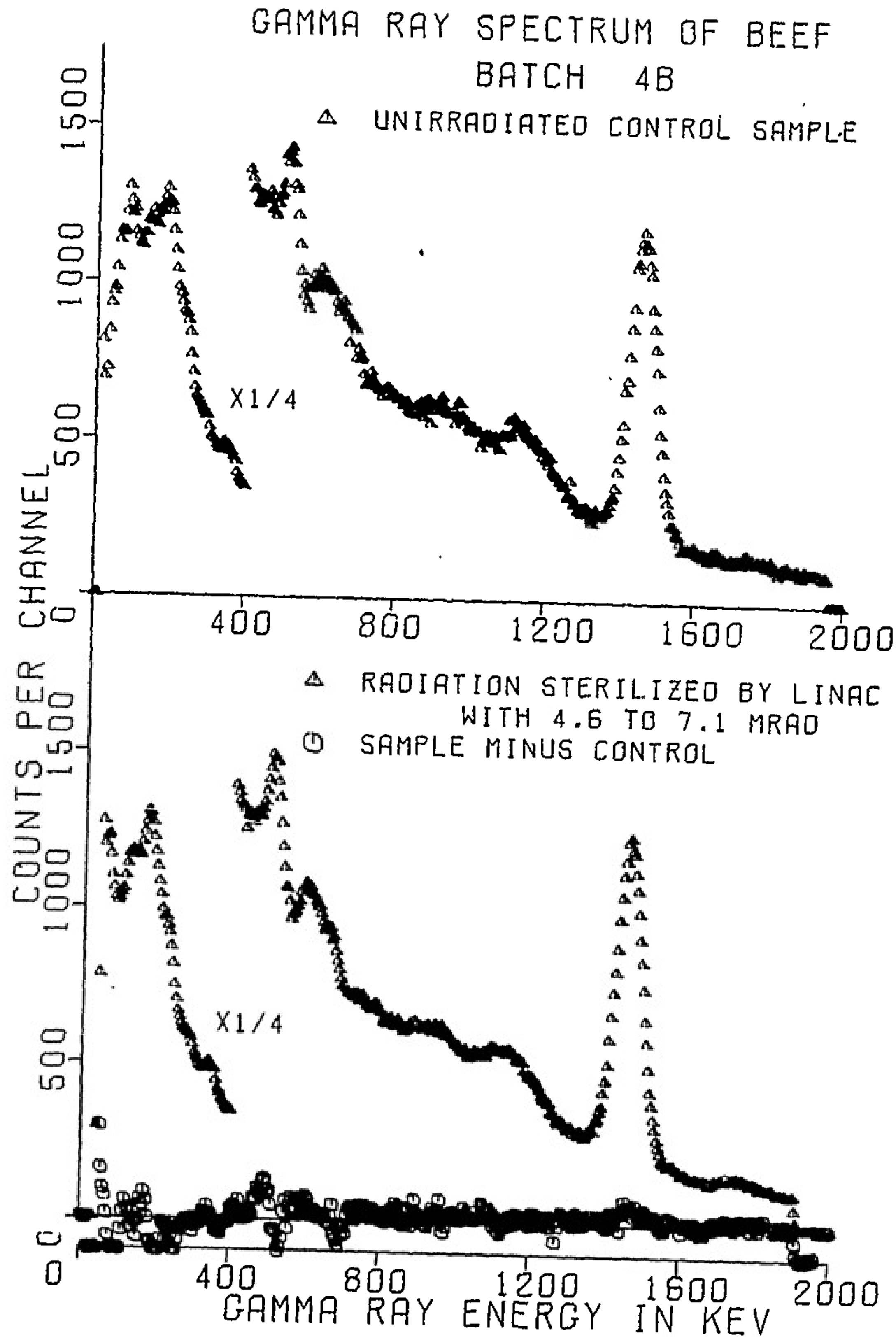
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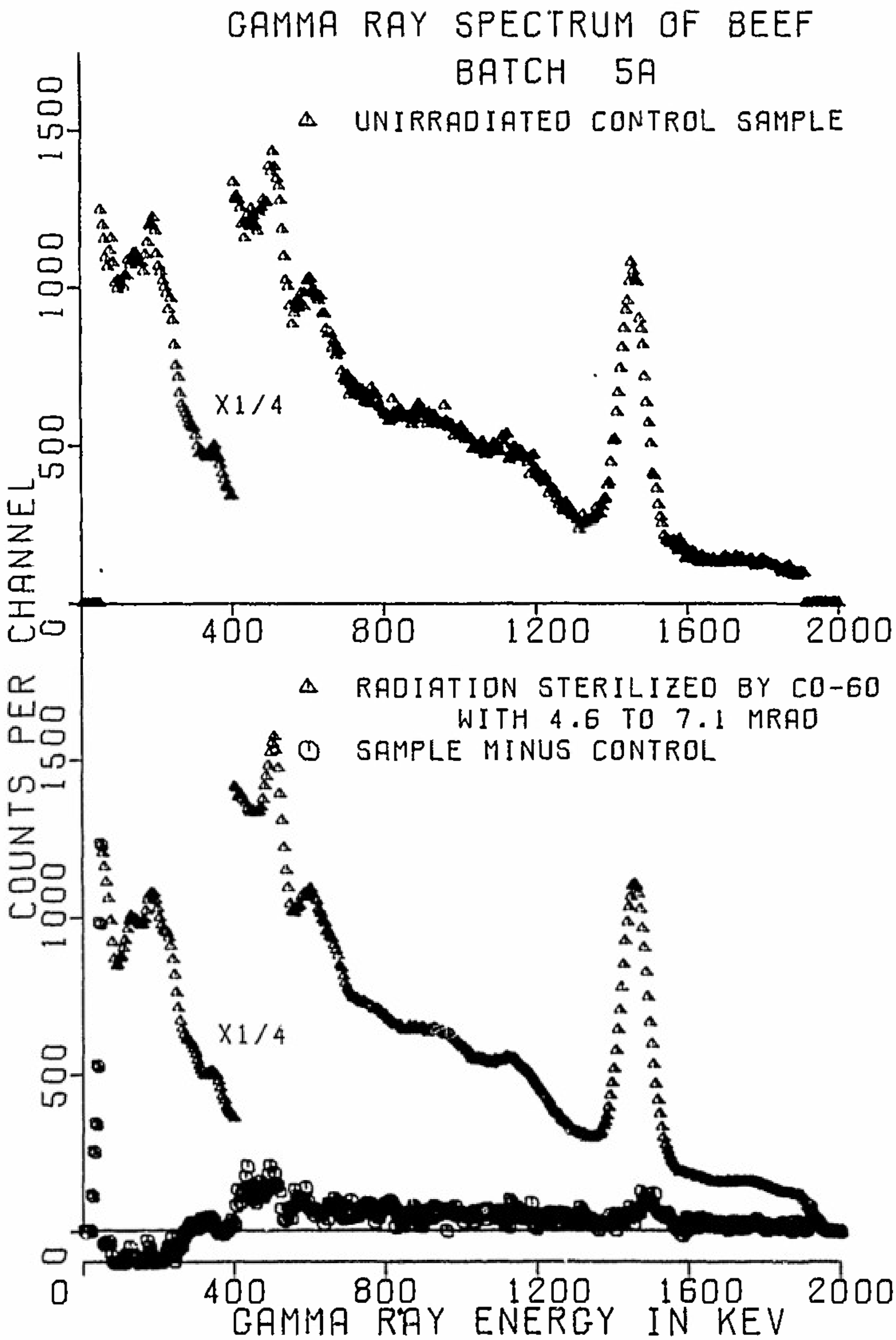
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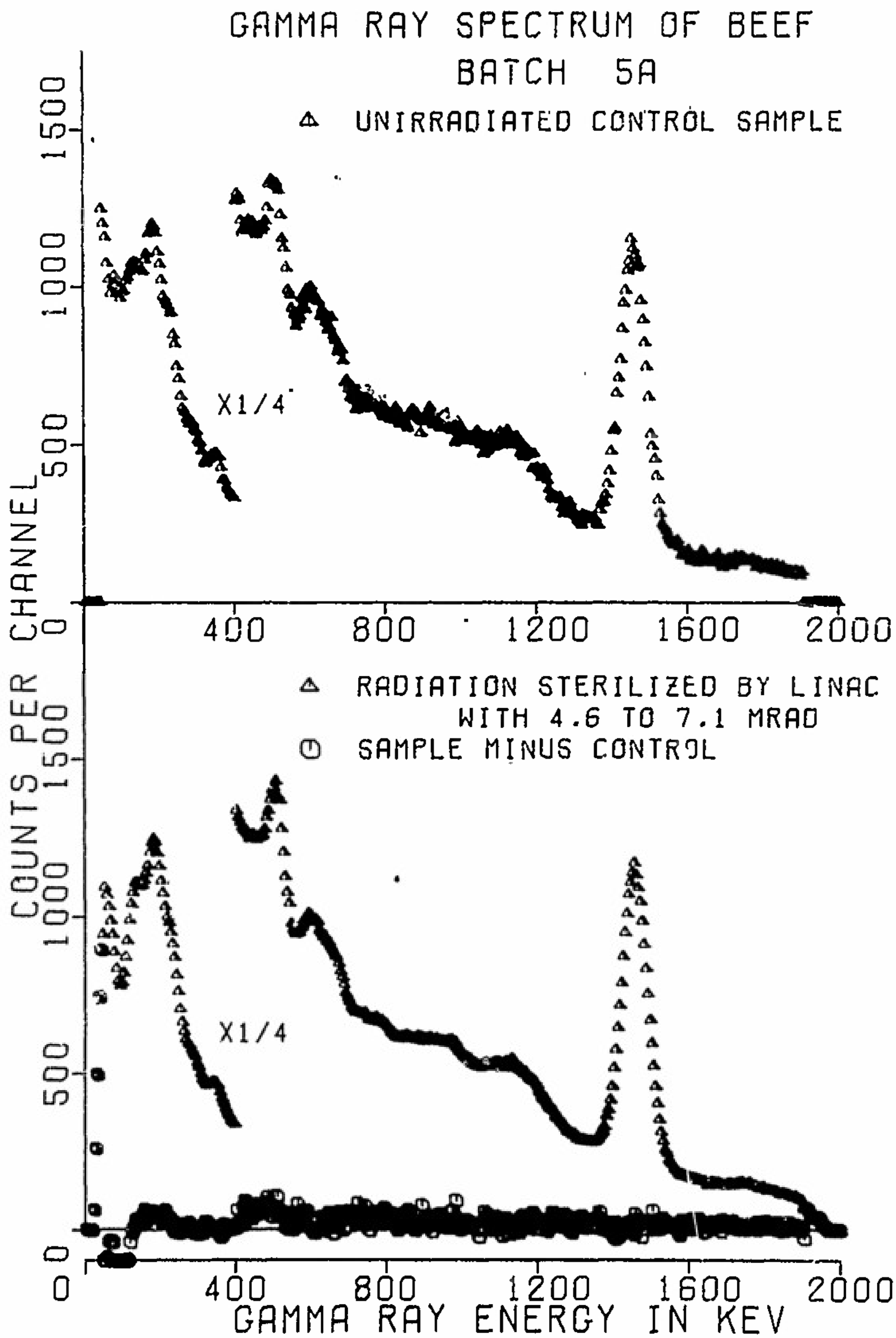
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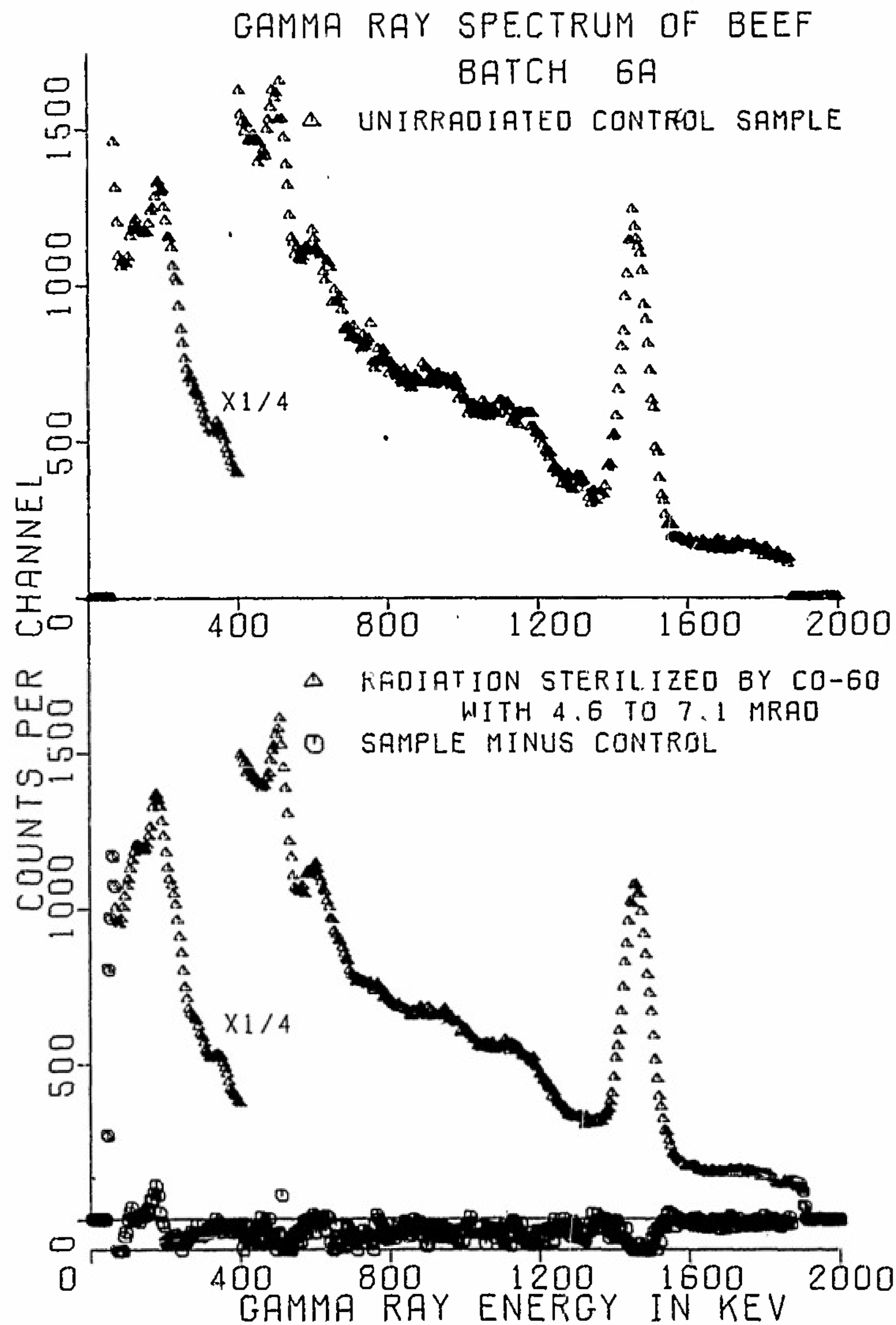
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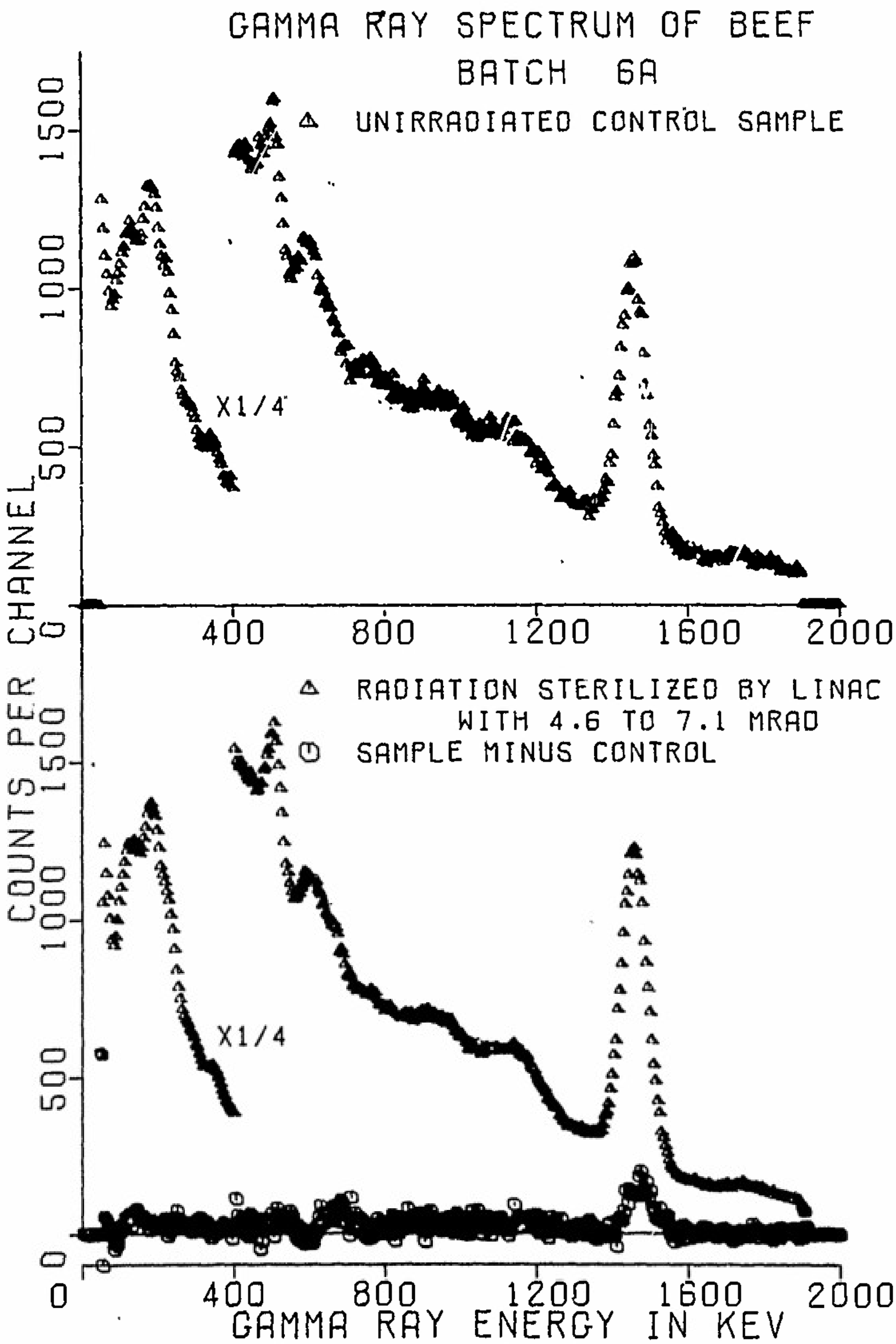
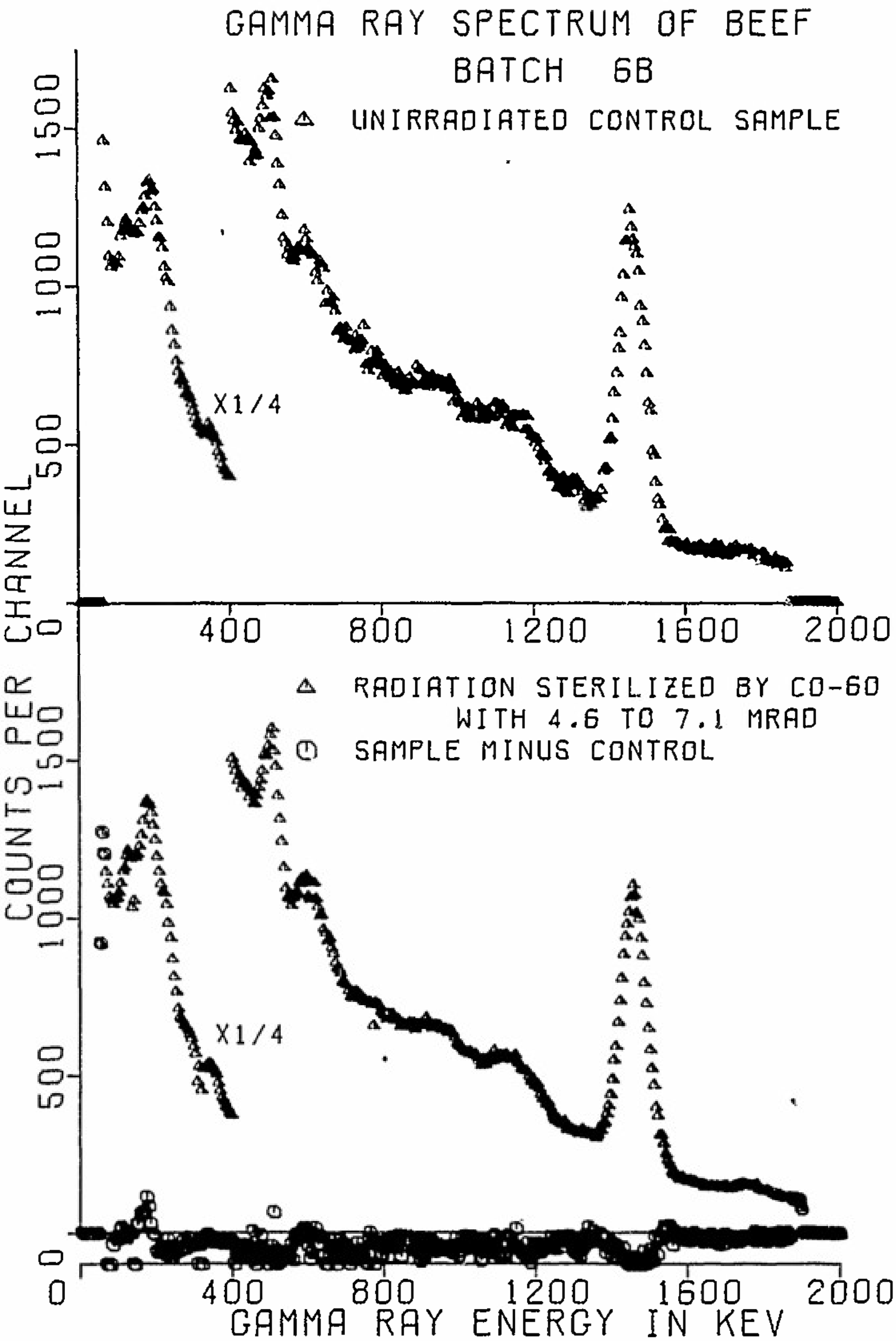
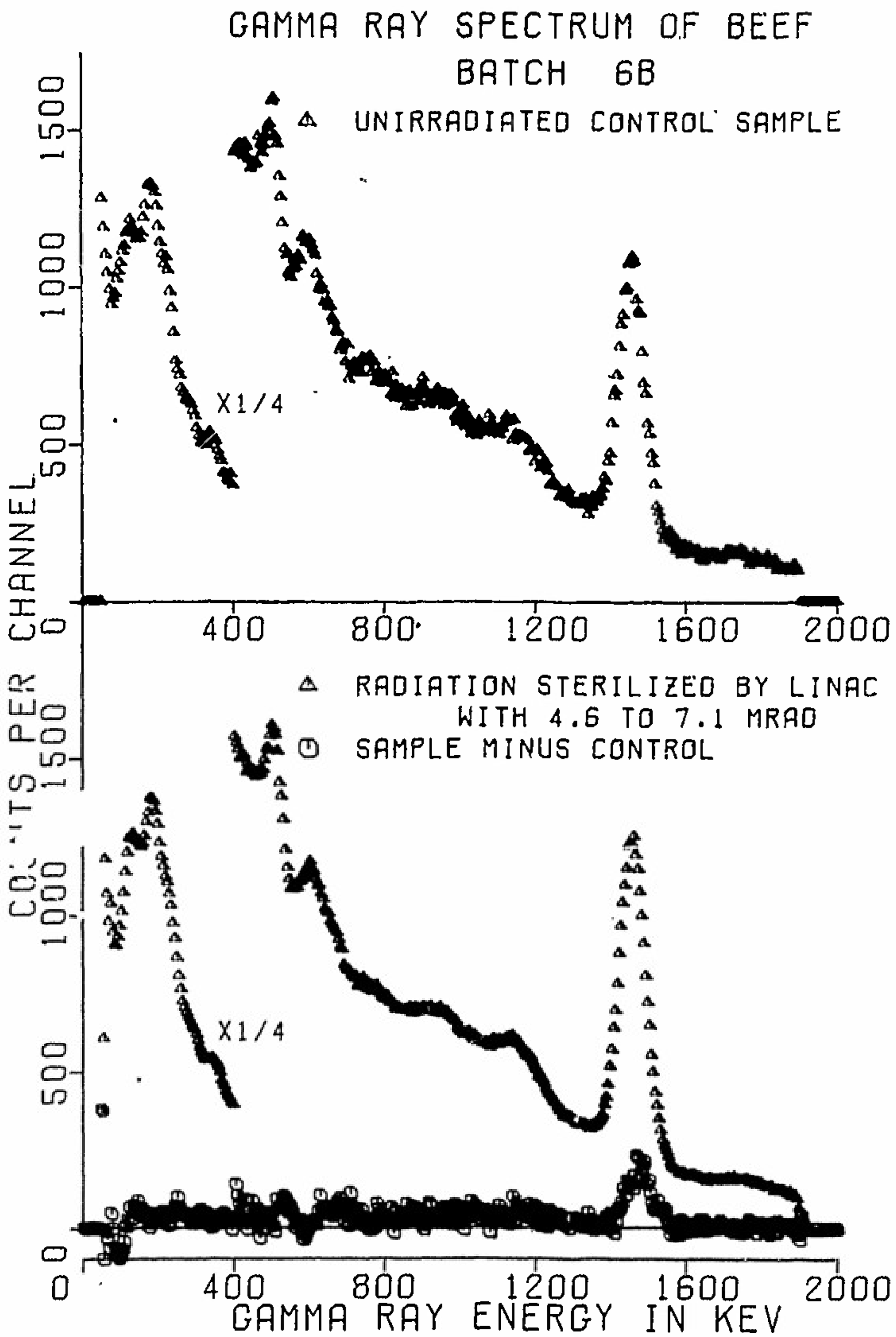
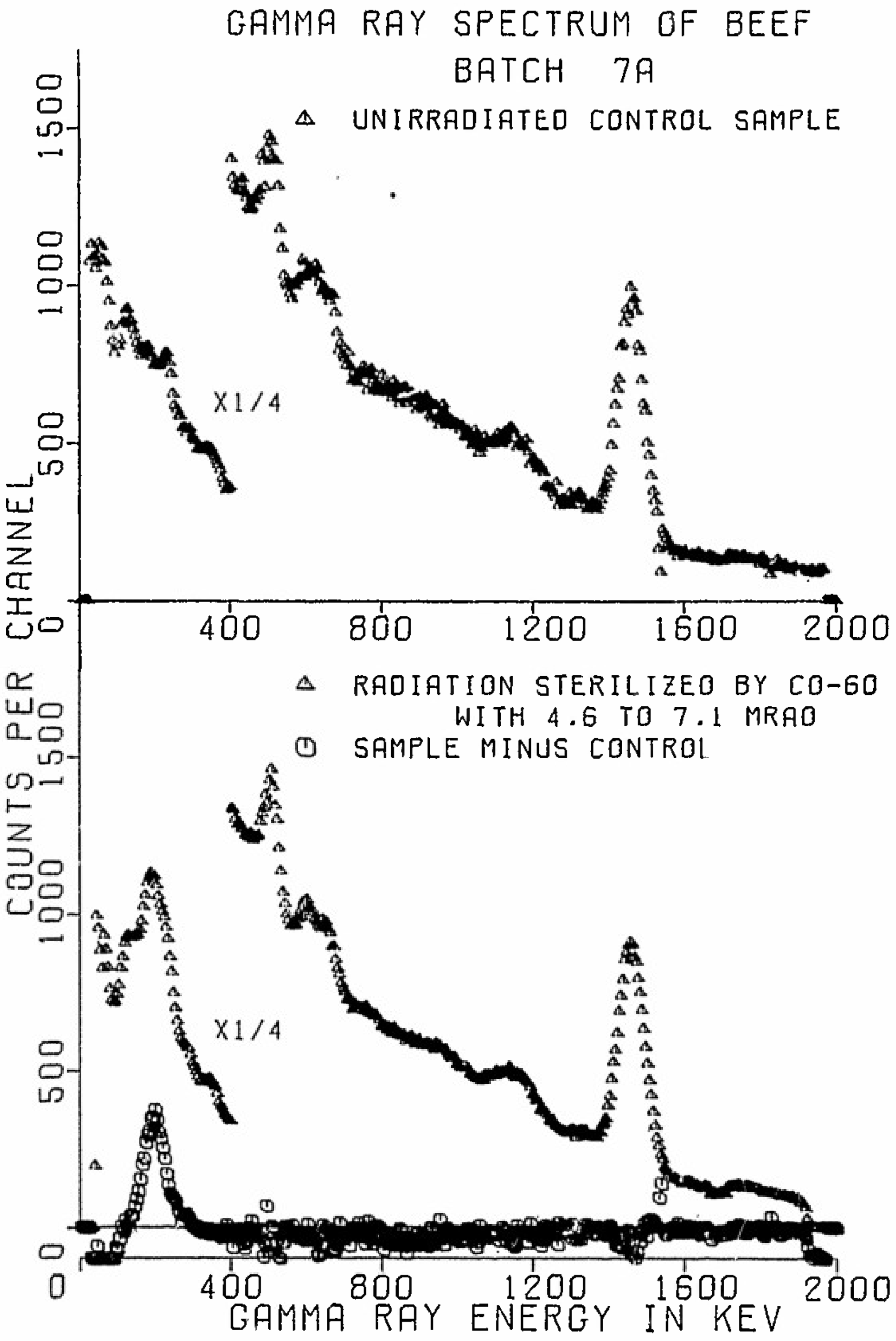
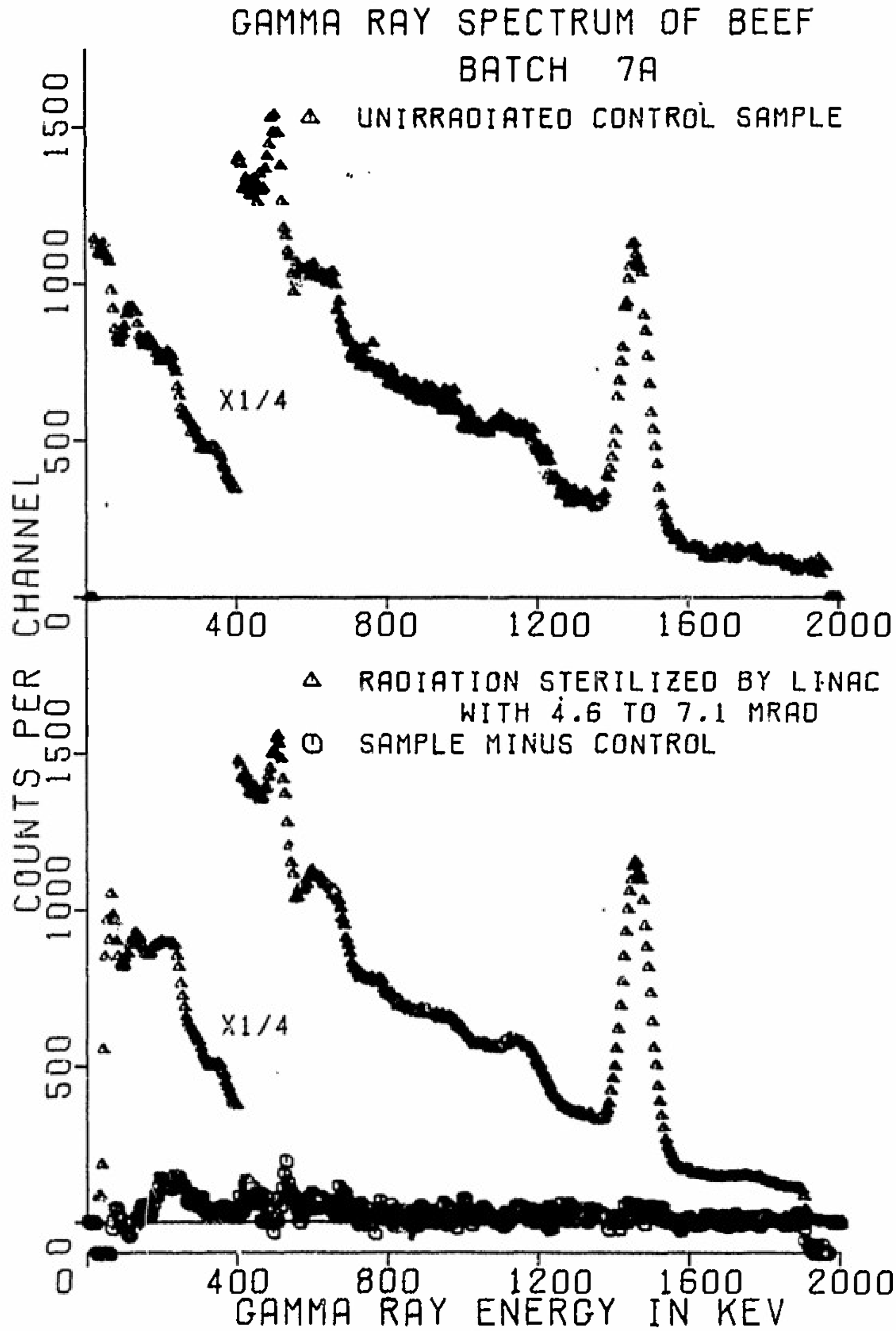

GAMMA RAY SPECTRUM OF BEEF
BATCH 6A
UNIRRADIATED CONTROL SAMPLE
X1/4
RADIATION STERILIZED BY LINAC
WITH 4.6 TO 7.1 MRAD
SAMPLE MINUS CONTROL
X1/4
COUNTS PER CHANNEL
0
500
1000
1500
400
800
1200
1600
2000
GAMMA RAY ENERGY IN KEV



GAMMA RAY SPECTRUM OF BEEF
BATCH 6B
UNIRRADIATED CONTROL SAMPLE
X1/4
RADIATION STERILIZED BY CO-60
WITH 4.6 TO 7.1 MRAD
SAMPLE MINUS CONTROL
X1/4
COUNTS PER CHANNEL
GAMMA RAY ENERGY IN KEV
0
500
1000
1500
400
800
1200
1600
2000


GAMMA RAY SPECTRUM OF BEEF
BATCH 6B
UNIRRADIATED CONTROL SAMPLE
X1/4
RADIATION STERILIZED BY LINAC
WITH 4.6 TO 7.1 MRAD
SAMPLE MINUS CONTROL
X1/4
COUNTS PER CHANNEL
0
500
1000
1500
0
400
800
1200
1600
2000
GAMMA RAY ENERGY IN KEV


GAMMA RAY SPECTRUM OF BEEF
BATCH 7A
UNIRRADIATED CONTROL SAMPLE
X1/4
RADIATION STERILIZED BY CO-60
WITH 4.6 TO 7.1 MRAD
SAMPLE MINUS CONTROL
X1/4
COUNTS PER CHANNEL
GAMMA RAY ENERGY IN KEV
0
500
1000
1500
400
800
1200
1600
2000


GAMMA RAY SPECTRUM OF BEEF
BATCH 7A
UNIRRADIATED CONTROL SAMPLE
X1/4
RADIATION STERILIZED BY LINAC
WITH 4.6 TO 7.1 MRAD
SAMPLE MINUS CONTROL
X1/4
COUNTS PER CHANNEL
GAMMA RAY ENERGY IN KEV
0
500
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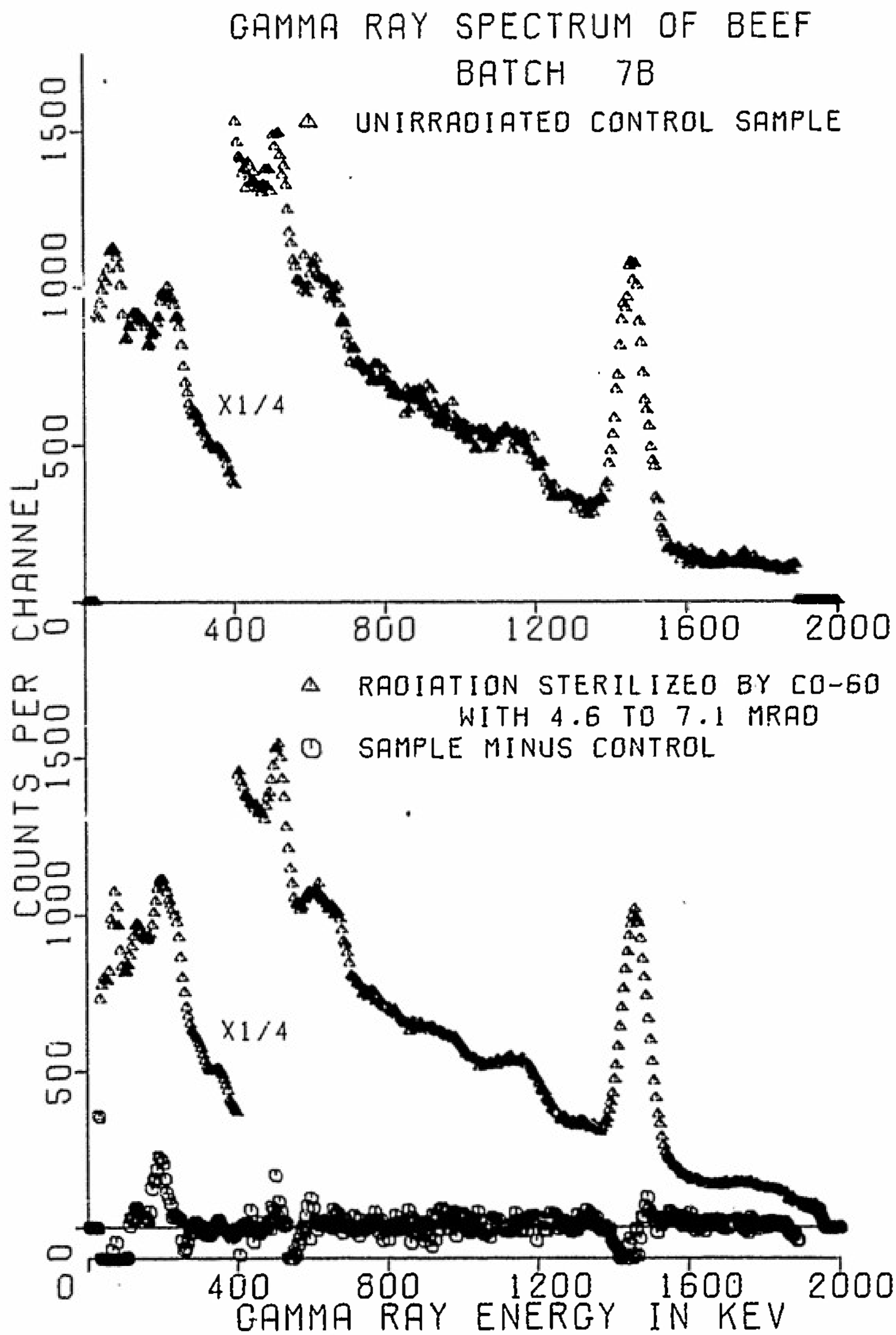
GAMMA RAY SPECTRUM OF BEEF
BATCH 7B
UNIRRADIATED CONTROL SAMPLE
X1/4
RADIATION STERILIZED BY CO-60
WITH 4.6 TO 7.1 MRAD
SAMPLE MINUS CONTROL
X1/4
COUNTS PER CHANNEL
GAMMA RAY ENERGY IN KEV
0
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1600
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APPENDIX B

Description of Computer Programs

Because of the massive amounts of data requiring complex processing, it was necessary to perform the analysis with the aid of a computer. A sequence of about ten computer codea was prepared for use in this work. These programs are described in this section.

CMEPTR,BCD controls the paper tape reader. Data were read from the paper tape in records of 119 characters each, and stored on magnetic tape with no change in the binary-coded-decimal format. Such records would normally contain ten spectrometer channels of data. There would be six digits to represent the contents, as well as a quadrant number and a channel number supplied for each channel of data. An end-of-record character, punched after every ten channels on the tape and read by the computer as a '#', would normally be the last character on each record, except where a character had been dropped or some other error had occurred in handling the data up to that point.

NLRLB1 transfers the data provided by the previoua program record by record, from the BCD code in which they were stored on the magnetic tape to a Fortran-readable form on the mass-storage of the Univac computer. At this point any redundancies caused by multiple reading of the same spectrum from paper tape were eliminated, so that there were 41 records of data for each spectrum.

NLRLB2 decodes the output provided by NLRLB1 so that the contents of each channel could be extracted. A spectrum is represented as a sequence of 400 6-digit numbers permitting the subsequent performance of numerical operations and other processing of the data. This reformatting of the data was accomplished easily except cases frequently encountered in which characters had been dropped or other errors had occurred. Such errors, usually noticed by a shift of the # symbol from its place at the end of a record were individually corrected.

NLRLB3 provides a permanent record of the raw data, with two pages of computer printout for each of the opectra. On one page the spectrum is printed, channel by channel. The data are headed by a compilation of relevant information such as the weight of the sample, the type of irradiation which had been given the sample, the irradiation date, the counting date, and the batch number of the sample. On the second page a plot of the spectrum is generated by the line printer of the computer.

NLRLB6 corrects certain data processing errors. Each of the more than 250 spectra, including more than 100,000 channels of information, were scanned by eye to identify data-processing errors that had been missed up to that point. Only a few of these were found to be present, and in every case, the error would involve only one channel, the content of which would be badly out of line with a smooth curve which would pass through all the neighboring points of the spectrum. Such errors, when found, were corrected to lie on the smooth curve consistent with the other points. This correction was necessary so that subsequent peak-search routines which were employed would not identify such a deviation as a peak from a radioactive decay.

NLRLB4 generates a plot on the line printer of regions of the pulse height spectrum in the vicinity of 511 and 1460 keV. These were the two energies at which clearly-identified gamma rays were present in the spectrum of each sample, from positron annihilation and from the decay of ^{40}K, respectively. The channel numbers at which these two peaks were observed were noted for each spectrum, and this provided a means of shifting the spectrum so that a consistent energy scale was available for all the data. The energy scale was made to be exactly 5 keV per channel. In cases where the energy scale was expanded or contracted, the number of counts in each channel were decreased or increased in the amount appropriate to keep the total counts constant.

NRLRB7 provides a backup data storage tape. The spectra, in the various stages in which they have been prepared at this point, were written onto a backup tape so that they could not be destroyed by a computer malfunction.

NLRLB8 averages all spectra within a particular batch for samples that had been subjected to the same type of irradiation. Such averaged spectra could be compared to control samples of the same batch which had not been irradiated, to determine the effect of the radiation in inducing radioactivity into the samples. Mass storage files were prepared containing averaged spectra, control spectra, and net counts containing the differences between the averaged and the control spectra, which were used for the routines described next.

NLRLB10 present line-printer plots of the various data files mentioned in the previous section, including averaged, control and net counts.

NLRLB5, NLRLB11 search the data for evidence of induced radioactivity. The individual spectra as well as the averaged spectra were subjected to a peak-search routine that attempted to find evidence for radioactivity, using curve-fitting procedures, that would not be apparent in a visual inspection of the data. While the peaks resulting from positron annihilation and ^{40}K, which were mentioned earlier in connection with the energy calibration, were observed in every sample, and occasionally there were very small peaks that appeared to be ^{137}Cs (apparently from fall-out from nuclear weapons testing) or from naturally-occurring radioactivity (thorium isotopes, for example), there was no indication of any radiation that could be attributed in any way to the effects of the irradiation by the LINAC or ^{60}Co sources.